FINITE AMPLITUDE CAPILLARY WAVES IN A LIQUID DROP

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ABSTRACT

A theoretical study of finite amplitude waves in an approximately spherical liquid drop subject to surface tension forces is presented. Equations inferred from hydrodynamic theory provide the starting point for the analysis. It is shown that the equations of motion for the liquid drop can be expressed in canonical form based on a certain postulated Hamiltonian. It is also shown that energy is a constant of the motion in this exact theory, but not in low order perturbation theory of an almost spherical drop. Exact expressions are expanded to obtain zeroth, first, and second order equations in perturbation theory. The zeroth order theory gives the Young-Laplace formula for a spherical drop. The first order equations are solved and the solutions are in agreement with Rayleigh's results for capillary waves with small amplitude. The second order equations take into account finite amplitudes and they contain the leading order nonlinear interactions among capillary waves. The complete, exact solutions of those equations for the velocity potential and surface displacement developed here are new and are a principal focus of this study. It is shown with the aid of these solutions that evolution of waves in time under resonance conditions can be produced to good approximation under near-resonance conditions provided that the interval of observation is suitably restricted. Further, it is shown that under near-resonance conditions there will be two or more frequencies of oscillation in any spatial state. These properties of the new formulas suggest a physical mechanism responsible for intermittency in a turbulent fluid. In the case of capillary ripple turbulence that mechanism involves a

certain kind of three-wave mixing. Experimental and theoretical implications of this work are discussed. On the theoretical side, the variational method used in establishing the canonical form of the equations of motion is explained in detail and compared and contrasted with a variational method used by others. Further, the importance of near-resonance terms in making a transition from a discrete to a continuous wave spectrum is discussed. Also, problems due to possible L=1 tesseral harmonic contributions to the second order perturbation formulas are identified. A planned NASA space flight experiment to study capillary ripple turbulence in an isolated liquid drop provided the principal motivation and direction for this research, and the new theoretical results will be available to aid in the analysis of observations made there. Results obtained here also provide new perspective for certain problems in ocean dynamics and plasma turbulence that involve high amplitude waves and nonlinear interactions. Finally, this research suggests experiments that could be performed to detect non-linear interactions among collective excitations in an atomic nucleus.

I. INTRODUCTION

Finite amplitude capillary waves in an almost spherical liquid drop are studied theoretically in this paper. Such waves provide an opportunity to investigate nonlinear interactions and their consequences in a relatively simple theory that can be subjected to controlled experimental tests under nearly ideal conditions. A highlight of current interest in this area is a planned NASA space flight experiment [1-3] to investigate capillary ripple turbulence in isolated drops.

Viewed in the general context of high amplitude waves and nonlinear interactions, this analysis bears on theories of such diverse phenomena as ocean dynamics,[4-9] plasma turbulence,[4,9] confinement of plasmas in controlled fusion,[9] and low energy nuclear physics.[10] A number of specific implications and features of the new results will be discussed. For example, recent observations[3] of intermittency in ripple wave turbulence have demonstrated that some degree of coherence still exists in turbulent systems. Other research has focused attention on the phenomenon of intermittency in fluid turbulence in a more general context, and the need for new approaches to understand it has been recognized.[11] A physical mechanism responsible for intermittent ordered structures in turbulence is suggested by the analysis presented here.

The theory is formulated in spherical coordinates and variables that are appropriate for the geometry of almost spherical drops. The starting point is a set of exact expressions for equations of motion of a perturbed liquid, boundary conditions, and initial conditions that was deduced initially from the hydrodynamic theory of an incompressible, inviscid liquid without vorticity, but with a moving surface subjected to forces associated with surface tension. An exact Hamiltonian is subsequently postulated and it is demonstrated with the aid of a variational method that the exact equations of motion can be expressed as Hamilton's canonical equations. It is then shown that in this exact theory, energy is a constant of the motion.

Perturbation equations and formulas are then derived thru expansions of exact expressions. The zeroth and first order versions of perturbation theory include known results[1 2] contained in the Young-Laplace formula for the pressure difference across the surface of a spherical drop, and in Rayleigh's linear theory of capillary waves in an almost spherical drop, respectively. Substitution of solutions of the linear equations of motion into the second order Hamiltonian reveals that in this level of approximation in spherical geometry, energy is not a constant of the motion. This result may be expected to lead to special problems in power spectrum characterization of ripple turbulence on a spherical surface, such as in the planned NASA space flight experiment. Similar problems do not occur in treating waves on almost flat surfaces.

The second order equations of motion contain the leading order nonlinear interactions among capillary waves. A principal focus of this paper is development of an exact solution of these second order equations. Both the phases and amplitudes of oscillating variables appear in the solution. Ensemble averages of the primary variables and related correlation functions could readily be calculated, if desired, in straightforward extensions of this work.

The dispersion curve for capillary waves bends upward in the linear theory in spherical geometry, just as in flat geometry. The upward bending implies that decay and coalescence processes can contribute to the second order equations of motion. This is an important simplifying feature in the analysis of time dependence in surface waves of the capillary type, as contrasted with gravitational waves in flat geometry. In the case of gravitational waves, decay does not occur in second order, but important time dependence associated with wave scattering, decay and coalescence occurs in third order perturbation theory. Third order perturbation theory is significantly more complex than second order theory.

The frequency spectrum for waves in the linear theory is discrete, and this discreteness is reflected in solutions of the second order equations of motion. A noteworthy feature of the exact solution of the second order equations is that the linear time growth of dynamic variables that occurs in the rare case of exact resonance can be produced to good approximation using Taylor's series expansions of near-resonance exact formulas, provided that the time interval of applicability is suitably restricted. The near-resonance condition occurs much more commonly than exact resonance, and the exact formulas, without expansion, contain important information for longer time intervals. Another important result of second order theory is that oscillations with multiple frequencies occur in any given spatial state.

Problems with making a proper transition from a discrete to a continuous wave spectrum in treating resonance and near-resonance conditions in nonlinear theories have been discussed by others.[13,14] Some difficult issues were encountered in treating near-resonance terms as well as other off-resonance terms as Cauchy principal values in integrals involving frequency differences. In some instances the principal value terms were neglected because they involve simple oscillatory behavior and were regarded as unimportant in understanding random distributions of weakly interacting waves. The results for time dependence in the present theory where the wave spectrum is definitely discrete bears on these issues and may aid in clarifying them.

Mathematical details and physical implications of the exact theory of capillary waves in a liquid drop are treated first in the following sections of this paper. Then the second order theory that includes the leading nonlinear interactions is developed in detail.

The discussion in the final section of this paper includes comments on the canonical formalism for a hydrodynamic system with a moving boundary. Methods used in this paper are compared and contrasted with earlier work of others.[15] The possibility of L=1 spherical harmonic contributions in the theory and problems associated with this are discussed. The physical mechanism responsible for intermittency suggested by the new results is explained. The relevance of the new theory to turbulence, like that in a stormy sea, on the surface of an atomic nucleus, and possibilities for observing footprints of this phenomenon in γ-ray spectra are discussed. Finally, experimental results[16] for large amplitude oscillations in levitated electrically charged liquid drops are cited in support of the predictions for behavior of a perturbed atomic nucleus.

II. DESCRIPTION OF MODEL AND FORMULATION OF AN EXACT THEORY

In the model for capillary waves in an isolated liquid drop considered here, it is assumed that no net external force acts on the drop and that the center of mass is stationary in an inertial reference frame.

The surface of the perturbed drop is at $r=r(\theta,\varphi;t)$, where θ and φ are spherical coordinates referred to an origin at the center of mass, and t is time. If $\eta(\theta,\varphi;t)$ is the radial displacement of the perturbed liquid surface from the unperturbed surface having radius r_0, then

$$r(\theta,\varphi;t) = r_0 + \eta(\theta,\varphi;t) \ . \tag{1}$$

In this analysis the liquid is regarded as incompressible and inviscid. Its density is ρ. It is assumed that only potential flow occurs, and at any point inside the drop or on its boundary the velocity $\vec{v}$ is given by

$$\vec{v}(r,\theta,\varphi;t) = \vec{\nabla}\Phi(r,\theta,\varphi;t) \, . \tag{2}$$

The following notation is used in what follows:

$$\frac{\partial\eta(\theta,\varphi;t)}{\partial t} = \dot{\eta} \, , \tag{3a}$$

$$\frac{\partial\Phi(r,\theta,\varphi;t)}{\partial t}=\dot{\Phi}\,. \tag{3b}$$

Hydrodynamic theory then provides a system of equations that determines Φ and η, as indicated below.

Continuity equation:

$$\nabla^2\Phi=0 \quad \text{for} \quad 0\le r\le r_0+\eta(\theta,\varphi;t)\,. \tag{4}$$

Kinematic boundary condition at the center of the drop:

$$\frac{\partial\Phi}{\partial r}=0 \quad \text{at} \quad r=0\,. \tag{5}$$

Kinematic boundary condition at the free surface:

$$\dot{\eta}-\left(\hat{r}-\vec{\nabla}\eta\right)\cdot\vec{\nabla}\Phi=0 \quad \text{at} \quad r=r_0+\eta(\theta,\varphi;t)\,, \tag{6}$$

where $\hat{r}$ is a unit vector in the radial direction.

Initial values of the free surface coordinate:

$$\eta(\theta,\varphi;t=0)=\eta_0(\theta,\varphi)\,. \tag{7}$$

Initial values of the free surface velocity:

$$\left[\hat{n}\cdot\vec{\nabla}\Phi(r,\theta,\varphi;t=0)\right]_{r=r_0+\eta(\theta,\varphi;t=0)}=\left[\hat{n}\cdot\vec{\nabla}\Phi_0(r,\theta,\varphi)\right]_{r=r_0+\eta_0(\theta,\varphi)}, \tag{8}$$

where $\hat{n}$ is the unit normal to the free surface.

Cauchy's integral for fluid motion:

$$\dot{\Phi}+\frac{1}{2}v^2+\frac{p}{\rho}=0 \quad \text{for} \quad 0\leq r\leq r_0+\eta(\theta,\varphi;t). \tag{9a}$$

A formula for the pressure p at any point just inside the free surface is derived in Appendix A. When that formula and Eq. (2) are inserted in Eq. (9a), one obtains the following result.

Dynamical boundary condition at the free surface:

$$\left\{\dot{\Phi}+\frac{1}{2}\left(\vec{\nabla}\Phi\right)^2+\frac{p_0}{\rho}+\frac{\alpha}{\rho}\frac{1}{r^2}\left\{r\left(\frac{1}{\gamma}+\gamma\right)-\frac{1}{\sin\theta}\left[\frac{\partial}{\partial\theta}\left(\frac{\sin\theta}{\gamma}\frac{\partial\eta}{\partial\theta}\right)\right]-\frac{1}{\sin^2\theta}\left[\frac{\partial}{\partial\varphi}\left(\frac{1}{\gamma}\frac{\partial\eta}{\partial\varphi}\right)\right]\right\}\right\}$$
$$=0 \qquad \text{at} \qquad r=r_0+\eta(\theta,\varphi;t). \tag{9b}$$

Equations (1)-(9b) provide a complete set of relations for an exact mathematical description of the model system.

Certain important properties of the exact solutions of these equations and results that are useful for perturbation theory can be exhibited by identifying the Hamiltonian and expressing Eqs. (6) and (9b) in Hamilton's canonical form. These basic expressions are treated next, in Sec.III.

III. THE HAMILTONIAN AND HAMILTON'S EQUATIONS

A. The Hamiltonian

A postulated Hamiltonian for the drop will be written as the sum of three terms, as follows:

$$H = H_a + H_b + H_c \ . \tag{10}$$

Kinetic energy, H_a:

$$H_a = \frac{1}{2}\rho \int_V d^3r \left(\vec{\nabla}\Phi\right)^2 \tag{11a}$$

$$= \frac{1}{2}\rho \int_0^{2\pi} \int_0^{\pi} \int_0^{r_0+\eta(\theta,\varphi;t)} (\vec{\nabla}\Phi)^2 \, r^2 \sin\theta \, dr \, d\theta \, d\varphi \ , \tag{11b}$$

where V is the volume of the drop.

Potential energy H_b associated with surface tension α:

$$H_b = \alpha \int_S df \tag{12a}$$

$$= \alpha \int_0^{2\pi} \int_0^{\pi} \left\{ \left[1 + \left(\vec{\nabla}\eta\right)^2 \right]^{1/2} r^2 \right\}_{r=r_0+\eta(\theta,\varphi;t)} \sin\theta \, d\theta \, d\varphi, \tag{12b}$$

where S is the free surface and df is a differential element of S.

Potential energy H_c associated with volume of the drop when p_0 is the pressure of the surrounding vapor:

$$H_c = p_0 \int_0^{2\pi} \int_0^{\pi} \int_0^{r_0+\eta(\theta,\varphi;t)} r^2 \sin\theta \, dr \, d\theta \, d\varphi . \tag{12c}$$

Equation (12b) is derived from differential geometry in Appendix A, where a quantity γ is defined as follows:

$$\gamma \equiv \left\{ 1 + \left[\vec{\nabla} \eta(\theta,\varphi;t) \right]^2 \right\}^{1/2} . \tag{13}$$

For the surface coordinates $\eta(\theta,\varphi;t)$ on the actual path, the integral that occurs in H_c is just the constant volume of the incompressible fluid. However for variations of η away from the actual path, the volume and in turn H_c may vary also, and H_c is needed to arrive at the Cauchy integral of an equation of motion in its usual form, Eq. (9b).

B. Hamilton's canonical equations

The canonical form of the equations of motion can be developed by regarding $\Phi(r,\theta,\varphi;t)$ and $\eta(\theta,\varphi;t)$ as fields that can be varied independently. However, these variations are subject to the conditions that on the actual path, Φ satisfies the Laplace equation, Eq. (4), throughout the interior of the drop and on its surface, and that Eq. (5) is satisfied on the actual path.

Then for the actual path, the relation

$$\vec{\nabla}\cdot\left[\left(\vec{\nabla}\Phi\right)\Phi\right]=\Phi\nabla^2\Phi+\left(\vec{\nabla}\Phi\right)^2 \tag{14}$$

and Gauss's theorem can be applied to express the kinetic energy in terms of quantities evaluated at the free surface, as follows:

$$H_a=\frac{1}{2}\rho\int_S d\vec{f}\cdot\Phi\vec{\nabla}\Phi\,. \tag{15}$$

Next we will calculate the change in H_a when Φ is varied from the actual path while η is held fixed on the actual path. To do this let $\Phi\to\Phi+\delta\Phi$ in Eq. (11a), and obtain the following result thru first order terms in $\delta\Phi$:

$$\delta H_a^{\Phi}=H_a(\Phi+\delta\Phi)-H_a(\Phi) \tag{16a}$$

$$=\rho\int d^3r\left(\vec{\nabla}\Phi\right)\cdot\left(\vec{\nabla}\delta\Phi\right). \tag{16b}$$

Use

$$\vec{\nabla}\cdot\left[\left(\vec{\nabla}\Phi\right)\delta\Phi\right]=\left(\nabla^2\Phi\right)\delta\Phi+\left(\vec{\nabla}\Phi\right)\cdot\left(\vec{\nabla}\delta\Phi\right) \tag{17}$$

and Gauss's theorem to express Eq.(16b) as

$$\delta H_a^{\Phi}=\rho\int_S d\vec{f}\cdot\left(\vec{\nabla}\Phi\right)\delta\Phi\,, \tag{18}$$

or

$$\delta H_a^{\Phi} = \rho \int_0^{2\pi} \int_0^{\pi} \left\{ \left[\hat{r}' - \vec{\nabla}' \eta(\theta', \varphi'; t) \right] \cdot \left(\vec{\nabla}' \Phi \right) (\delta \Phi) (r')^2 \right\}_{r' = r_0 + \eta(\theta', \varphi'; t)} \sin\theta' \, d\theta' \, d\varphi' , \tag{19}$$

where the result for $d\vec{f}$ derived in Appendix A has been used. A basic variational derivative on the surface where $r = r_0 + \eta(\theta, \varphi; t)$ and $r' = r_0 + \eta(\theta', \varphi'; t)$ takes the form

$$\frac{\delta \Phi(r', \theta', \varphi'; t)}{\delta \Phi(r, \theta, \varphi; t)} = \frac{1}{r^2 \sin\theta} \delta(\theta - \theta') \delta(\varphi - \varphi') , \tag{20}$$

and this can be used to calculate the variational derivative $\delta H_a / \delta \Phi$ from Eq.(19). After integrating over δ functions and dividing by ρ, one finds the following result:

$$\left. \frac{1}{\rho} \frac{\delta H_a}{\delta \Phi(r, \theta, \varphi; t)} \right|_{r = r_0 + \eta(\theta, \varphi; t)} = \left\{ \left[\hat{r} - \vec{\nabla} \eta(\theta, \varphi; t) \right] \cdot \vec{\nabla} \Phi(r, \theta, \varphi; t) \right\}_{r = r_0 + \eta(\theta, \varphi; t)} . \tag{21a}$$

Note that

$$\frac{\delta H}{\delta \Phi} = \frac{\delta H_a}{\delta \Phi} \tag{21b}$$

because H_b and H_c do not involve Φ.

Let the canonical momentum π be defined by

$$\pi(r,\theta,\varphi;t)\big|_{r=r_0+\eta(\theta,\varphi;t)} = \rho\Phi(r,\theta,\varphi;t)\big|_{r=r_0+\eta(\theta,\varphi;t)}. \tag{22}$$

The notation in Eq. (22) is used to indicate that functions π and Φ are defined throughout the drop and on its surface and for all times. However, only when π is evaluated on the free surface is it the canonical momentum. Also, let the canonical coordinate variable be $\eta(\theta,\varphi;t)$. Then one of Hamilton's canonical equations for a continuum takes the form

$$\dot{\eta} = \frac{\delta H}{\delta \pi}. \tag{23}$$

Combining Eqs. (21a)-(23), one arrives at the following result

$$\dot{\eta}(\theta,\varphi;t) = \left\{ \left[\hat{r} - \vec{\nabla}\eta(\theta,\varphi;t)\right] \cdot \vec{\nabla}\Phi(r,\theta,\varphi;t) \right\}_{r=r_0+\eta(\theta,\varphi;t)}, \tag{24}$$

which is equivalent to the kinematic boundary condition in Eq.(6).

Consider next the first order variation δH^{η} of the Hamiltonian when Φ is held constant on the actual path and η is varied from the actual path by an amount $\delta\eta$ according to the prescription $\eta \rightarrow \eta + \delta\eta$. First treat δH_a^{η}. Variation of Eq. (11b) involves taking the derivative of the integral with respect to its upper limit. One obtains

$$\delta H_a^{\eta} = \frac{1}{2}\rho \int_0^{2\pi}\int_0^{\pi} \left[\left(\vec{\nabla}'\Phi\right)^2 (r')^2 \delta\eta \right]_{r'=r_0+\eta(\theta',\varphi';t)} \sin\theta' \, d\theta' \, d\varphi'. \tag{25}$$

A basic variational derivative on the surface where $r = r_0 + \eta(\theta, \varphi; t)$ and $r' = r_0 + \eta(\theta', \varphi'; t)$ takes the form

$$\frac{\delta\eta(\theta',\varphi';t)}{\delta\eta(\theta,\varphi;t)} = \frac{1}{r^2 \sin\theta}\delta(\theta-\theta')\delta(\varphi-\varphi')\,. \tag{26}$$

Equation (26) can be used to calculate the variational derivative $\delta H_b / \delta\eta$ from Eq. (25). After integrating over δ functions, one finds

$$\frac{\delta Ha}{\delta\eta(\theta,\varphi;t)} = \frac{1}{2}\rho\left(\vec{\nabla}\Phi\right)^2\Big|_{r=r_0+\eta(\theta,\varphi;t)}\,. \tag{27}$$

The variational derivatives $\delta H_b / \delta\eta$ and $\delta H_c / \delta\eta$ are evaluated in Appendix A, in Eqs. (A12) and (A15), respectively. The second of Hamilton's equations for a continuum is

$$\dot{\pi} = -\frac{\delta H}{\delta\eta}\,. \tag{28}$$

Combining Eqs. (10), (27), (A12), (A15), and (28), one finds

$$\rho\dot{\Phi}(r,\theta,\varphi;t)\Big|_{r=r_0+\eta(\theta,\varphi;t)} = \Bigg\{\frac{1}{2}\rho\left(\vec{\nabla}\Phi\right)^2$$

$$+\alpha\left(\frac{1}{r}\left(\frac{1}{\gamma}+\gamma\right)-\frac{1}{r^2\sin\theta}\left[\frac{\partial}{\partial\theta}\left(\frac{\sin\theta}{\gamma}\frac{\partial\eta}{\partial\theta}\right)\right]-\frac{1}{r^2\sin^2\theta}\left[\frac{\partial}{\partial\varphi}\left(\frac{1}{\gamma}\frac{\partial\eta}{\partial\varphi}\right)\right]\right) \tag{29}$$

$$+p_0\Bigg\}_{r=r_0+\eta(\theta,\varphi;t)} .$$

Simple algebraic rearrangement of Eq. (29) yields Eq. (9b), the dynamical boundary condition at the free surface.

C. Time derivative of the total energy

The Hamiltonian in Eq. (10) represents the total energy of the system, and the time derivative of the total energy can be readily evaluated.

First calculate dH_a/dt, as follows:

$$\frac{dH_a}{dt} = \frac{1}{2}\rho\Bigg\{\int_0^{2\pi}\int_0^{\pi}\int_0^{r_0+\eta(\theta,\varphi;t)} 2\left(\vec{\nabla}\Phi\right)\cdot\left(\vec{\nabla}\dot{\Phi}\right)r^2\sin\theta\,dr\,d\theta\,d\varphi$$

$$+\int_0^{2\pi}\int_0^{\pi}\left[\left(\vec{\nabla}\Phi\right)^2 r^2\dot{\eta}(\theta,\varphi;t)\right]_{r=r_0+\eta(\theta,\varphi;t)}\sin\theta\,d\theta\,d\varphi \tag{30a}$$

$$=\frac{dH_{a1}}{dt}+\frac{dH_{a2}}{dt}\Bigg\}. \tag{30b}$$

Next apply steps similar to those used in arriving at Eq.(19), but replace $\delta\Phi$ there by $\dot{\Phi}$. This yields the following result for dH_{a1}/dt::

$$\frac{dH_{a1}}{dt} = \rho \int_0^{2\pi} \int_0^{\pi} \int_0^{r_0+\eta(\theta,\varphi;t)} \left(\vec{\nabla}\Phi\right)\cdot\left(\vec{\nabla}\dot{\Phi}\right) r^2 \sin\theta \, dr \, d\theta \, d\varphi \tag{31a}$$

$$= \rho \int_0^{2\pi} \int_0^{\pi} \left\{ \left(\left[\hat{r} - \vec{\nabla}\eta(\theta,\varphi;t) \right] \cdot \vec{\nabla}\Phi(r,\theta,\varphi;t) \right) \dot{\Phi}(r,\theta,\varphi;t) r^2 \right\}_{r=r_0+\eta(\theta,\varphi;t)} \sin\theta \, d\theta \, d\varphi \tag{31b}$$

$$= \rho \int_0^{2\pi} \int_0^{\pi} \left\{ \dot{\eta}(\theta,\varphi;t) \dot{\Phi}(r,\theta,\varphi;t) r^2 \right\}_{r=r_0+\eta(\theta,\varphi;t)} \sin\theta \, d\theta \, d\varphi \, , \tag{31c}$$

where Eq. (24) was used in the last step above.

Next we will evaluate dH_{a2}/dt. With the aid of Eq. (27), this term can be expressed as

$$\frac{dH_{a2}}{dt} = \int_0^{2\pi} \int_0^{\pi} \left\{ \frac{\delta H_a}{\delta\eta(\theta,\varphi;t)} \dot{\eta}(\theta,\varphi;t) r^2 \right\}_{r=r_0+\eta(\theta,\varphi;t)} \sin\theta \, d\theta \, d\varphi \, . \tag{31d}$$

Now refer to Eq. (12b) and evaluate dH_b/dt. If one follows steps parallel to those used in deriving Eq. (A10) in Appendix A but replaces $\delta\eta$ there by $\dot{\eta}$ and then uses Eq. (A12), one obtains

$$\frac{dH_b}{dt} = \int_0^{2\pi} \int_0^{\pi} \left\{ \frac{\delta H_b}{\delta\eta(\theta,\varphi;t)} \dot{\eta}(\theta,\varphi;t) r^2 \right\}_{r=r_0+\eta(\theta,\varphi;t)} \sin\theta \, d\theta \, d\varphi \, . \tag{32}$$

Next refer to H_c in Eq. (12c) and evaluate dH_c/dt.. One finds

$$\frac{dH_c}{dt} = p_0 \int_0^{2\pi} \int_0^{\pi} \left\{\dot{\eta}(\theta,\varphi;t) r^2\right\}_{r=r_0+\eta(\theta,\varphi;t)} \sin\theta \, d\theta \, d\varphi \,. \tag{33}$$

The result in Eq. (A15) can be used to write Eq. (33) as

$$\frac{dH_c}{dt} = \int_0^{2\pi} \int_0^{\pi} \left\{ \frac{\delta H_c}{\delta\eta(\theta,\varphi;t)} \dot{\eta}(\theta,\varphi;t) r^2 \right\}_{r=r_0+\eta(\theta,\varphi;t)} \sin\theta \, d\theta \, d\varphi \,. \tag{34}$$

Combining the results in Eqs. (22), (28), and (31d)-(33), one finds

$$\frac{d}{dt}\left[H_{a2} + H_b + H_c\right] = \int_0^{2\pi} \int_0^{\pi} \left\{ \frac{\delta\left[H_a + H_b + H_c\right]}{\delta\eta(\theta,\varphi;t)} \dot{\eta}(\theta,\varphi;t) r^2 \right\}_{r=r_0+\eta(\theta,\varphi;t)} \sin\theta \, d\theta \, d\varphi \,. \tag{35a}$$

$$= \int_0^{2\pi} \int_0^{\pi} \left\{ -\rho\dot{\Phi}(r,\theta,\varphi;t)\dot{\eta}(\theta,\varphi;t) r^2 \right\}_{r=r_0+\eta(\theta,\varphi;t)} \sin\theta \, d\theta \, d\varphi \,. \tag{35b}$$

Finally, Eqs. (31c) and (35b) can be combined to obtain

$$\frac{dH}{dt} = \int_0^{2\pi} \int_0^{\pi} \left\{ \left[\rho\dot{\eta}\dot{\Phi} - \rho\dot{\Phi}\dot{\eta}\right] r^2 \right\}_{r=r_0+\eta(\theta,\varphi;t)} \sin\theta \, d\theta \, d\varphi = 0 \,, \tag{36}$$

Equation (36) shows that the total energy of the system is a constant of the motion in this exact theory. The importance of this result will become more evident later when a contrary result is reached in low order perturbation theory.

IV. PERTURBATION THEORY EXPANSIONS

Formulas for perturbation approximations can be conveniently generated in two stages. First, terms in the exact equations are expanded in terms of η about the radius r_0 of the quiescent free surface. In the next stage, Φ and η are expanded in powers of a small perturbation parameter.

A. Expansions about the radius r_0

Refer to Eq. (6) and expand components of the second term as follows:

$$\left(\hat{r}\cdot\vec{\nabla}\Phi\right)_{r=r_0+\eta(\theta,\varphi;t)}=\left(\frac{\partial\Phi}{\partial r}\right)_{r=r_0+\eta(\theta,\varphi;t)}=\left(\frac{\partial\Phi}{\partial r}\right)_{r_0}+\left(\frac{\partial^2\Phi}{\partial r^2}\right)_{r_0}\eta+\frac{1}{2!}\left(\frac{\partial^3\Phi}{\partial r^3}\right)_{r_0}\eta^2+\cdots \tag{37}$$

$$\left[\left(\vec{\nabla}\eta\right)\cdot\left(\vec{\nabla}\Phi\right)\right]_{r=r_0+\eta(\theta,\varphi;t)}$$
$$=\left[\left(\vec{\nabla}\eta\right)\cdot\left(\vec{\nabla}\Phi\right)\right]_{r_0}+\left[\frac{\partial}{\partial r}\left(\left(\vec{\nabla}\eta\right)\cdot\left(\vec{\nabla}\Phi\right)\right)\right]_{r_0}\eta+\frac{1}{2!}\left[\frac{\partial^2}{\partial r^2}\left(\left(\vec{\nabla}\eta\right)\cdot\left(\vec{\nabla}\Phi\right)\right)\right]_{r_0}\eta^2+\cdots. \tag{38}$$

An approximate expression for Eq. (6) sufficient for present purposes can then be found by substituting Eqs. (36) and (37) into Eq. (6) and retaining only terms that are accurate thru $O(\lambda^2)$. Here λ is a parameter that indicates the degree of nonlinearity and it represents a factor of Φ or η. This yields the following result.

Kinematic boundary condition at free surface accurate thru $O(\lambda^2)$:

$$\dot{\eta}-\left[\left(\frac{\partial\Phi}{\partial r}\right)_{r_0}+\left(\frac{\partial^2\Phi}{\partial r^2}\right)_{r_0}\eta\right]+\left[(\vec{\nabla}\eta)\cdot(\vec{\nabla}\Phi)\right]_{r_0}=0. \tag{39}$$

Turn next to expansion of Eq. (9b) about r_0. Note that p_0/ρ is a constant that does not depend on time or space coordinates. Expanding $\dot{\Phi}$ about r_0 , one finds:

$$\dot{\Phi}(r,\theta,\varphi;t)=\dot{\Phi}(r_0,\theta,\varphi;t)+\left(\frac{\partial}{\partial r}\dot{\Phi}\right)_{r_0}\eta+\frac{1}{2!}\left(\frac{\partial^2}{\partial r^2}\dot{\Phi}\right)_{r_0}\eta^2+\cdots \tag{40}$$

Also, $\left[\left(\vec{\nabla}\Phi\right)^2\right]_{r=r_0+\eta(\theta,\varphi;t)}$ and $\left(\frac{1}{r}\right)_{r=r_0+\eta(\theta,\varphi;t)}$ may be similarly expanded about r_0. Refer to Eqs. (A6a) and (A6d) and note that

$$\left[\left(\vec{\nabla}\eta\right)^2\right]_{r=r_0+\eta}=\left(\frac{1}{r^2}\right)_{r=r_0+\eta}\left[\left(\frac{\partial\eta}{\partial\theta}\right)^2+\frac{1}{\sin^2\theta}\left(\frac{\partial\eta}{\partial\varphi}\right)^2\right]. \tag{41}$$

$$\left[\gamma(r,\theta,\varphi;t)\right]_{r=r_0+\eta}=\left[1+\frac{1}{2}\left(\vec{\nabla}\eta\right)^2-\frac{1}{8}\left(\vec{\nabla}\eta\right)^4+\cdots\right]_{r=r_0+\eta} \tag{42a}$$

$$\left[\frac{1}{\gamma(r,\theta,\varphi;t)}\right]_{r=r_0+\eta}=\left[1-\frac{1}{2}\left(\vec{\nabla}\eta\right)^2+\frac{3}{8}\left(\vec{\nabla}\eta\right)^4+\cdots\right]_{r=r_0+\eta}. \tag{42b}$$

Using Eq. (42a) and (42b), one obtains

$$\left[\frac{1}{\gamma}+\gamma\right]_{r=r_0+\eta}=2+O\left(\eta^4\right). \tag{42c}$$

Substituting Eqs. (40)-(42c) into Eq. (9b), one finds the following result.

Dynamical boundary condition at the free surface, accurate thru $O(\lambda^2)$:

$$\begin{aligned}&\left\{[\dot{\Phi}]_{r_0}+\left[\left(\frac{\partial}{\partial r}\dot{\Phi}\right)\right]_{r_0}\eta+\frac{1}{2}\left[\left(\bar{\nabla}\Phi\right)^2\right]_{r_0}+\frac{p_0}{\rho}+\frac{\alpha}{\rho}\left[\frac{2}{r_0}-\frac{2\eta}{r_0^2}-\frac{1}{r_0^2}\left(\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial\eta}{\partial\theta}\right)\right.\right.\right.\\&\left.\left.\left.+\frac{1}{\sin^2\theta}\frac{\partial^2\eta}{\partial\varphi^2}\right)+\frac{2\eta^2}{r_0^3}+\frac{2\eta}{r_0^3}\left(\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial\eta}{\partial\theta}\right)+\frac{1}{\sin^2\theta}\frac{\partial^2\eta}{\partial\varphi^2}\right)\right]\right\}=0.\end{aligned} \tag{43}$$

B. Expansions in a perturbation parameter

The second stage in generating perturbation formulas involves expanding Φ and η in the form

$$\Phi(\bar{r};t)={}_0\Phi(t)+{}_1\Phi(\bar{r};t)+{}_2\Phi(\bar{r};t)+\cdots \tag{44a}$$

$$\eta(\theta,\varphi;t)={}_1\eta(\theta,\varphi;t)+{}_2\eta(\theta,\varphi;t)+\cdots, \tag{44b}$$

where a perturbation parameter has been drawn into the perturbation functions. The subscript indicates the order of magnitude of a function. The linear equations (4) and (5) then yield

$$\nabla^2{}_n\Phi=0 \quad for \quad 0\le r\le r_0+\eta, \tag{45a}$$

$$\nabla^2{}_n\Phi=0 \quad for \quad 0\le r\le r_0+\eta \ . \tag{45a}$$

By substituting Eqs. (44a) and (44b) into Eq. (9), one obtains the following kinematic boundary conditions for the perturbation functions, evaluated at r_0:

$$ {}_1\dot{\eta} - \left(\frac{\partial_1\Phi}{\partial r}\right)_{r_0} = 0 \tag{46a} $$

$$ {}_2\dot{\eta} - \left(\frac{\partial_2\Phi}{\partial r}\right)_{r_0} - \left(\frac{\partial^2{}_1\Phi}{\partial r^2}\right)_{r_0} {}_1\eta + \left[\left(\bar{\nabla}_1\eta\right)\cdot\left(\bar{\nabla}_1\Phi\right)\right]_{r_0} = 0\,. \tag{46b} $$

Substituting Eqs. (44a) and (44b) into Eq. (43), one obtains the following dynamic boundary conditions for the perturbation functions, evaluated at r_0:

$$ \left({}_0\dot{\Phi}\right)_{r_0} + \frac{p_0}{\rho} + \frac{2\alpha}{\rho r_0} = 0 \tag{47a} $$

$$ \left({}_1\dot{\Phi}\right)_{r_0} - \frac{\alpha}{\rho r_0{}^2}\left[2{}_1\eta + \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial_1\eta}{\partial\theta}\right) + \frac{1}{\sin^2\theta}\frac{\partial^2{}_1\eta}{\partial\varphi^2}\right] = 0 \tag{47b} $$

$$ \begin{aligned} &\Bigg\{\left({}_2\dot{\Phi}\right)_{r_0} + \left(\frac{\partial}{\partial r}{}_1\dot{\Phi}\right)_{r_0} {}_1\eta + \frac{1}{2}\left[\left(\bar{\nabla}_1\Phi\right)^2\right]_{r_0} - \frac{\alpha}{\rho r_0{}^2}\Bigg[2{}_2\eta + \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial_2\eta}{\partial\theta}\right) + \frac{1}{\sin^2\theta}\frac{\partial^2{}_2\eta}{\partial\varphi^2} \\ &\qquad - \frac{2{}_1\eta}{r_0}\left({}_1\eta + \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial_1\eta}{\partial\theta}\right) + \frac{1}{\sin^2\theta}\frac{\partial^2{}_1\eta}{\partial\varphi^2}\right)\Bigg]\Bigg\} = 0\,. \end{aligned} \tag{47c} $$

The zeroth order equation, Eq. (47a), yields the Young-Laplace formula[12] for the case of an undisturbed spherical drop in its usual form if one writes

$$\left({}_0\Phi\right)_{r_0} = -\frac{p_1}{\rho}t, \tag{48a}$$

where p_1 is the constant pressure just inside the free surface. Then Eq. (47a) can be expressed as

$$p_1 - p_2 = \frac{2\alpha}{r_0}. \tag{48b}$$

C. Energy perturbation thru second order terms

The goal here is to develop a formula for energy that takes into account the leading order terms that depend on the solutions of the linearized equations of motion, ${}_1\Phi$ and ${}_1\eta$, and neglects all nonlinear effects involving ${}_2\Phi$ and ${}_2\eta$ and higher order terms. Energy terms involving ${}_1\Phi$ and ${}_1\eta$ only thru second order will be retained. Perturbative formulas for energy will be derived here by expansion of terms H_a and H_b.. The external pressure p_0 is unimportant for this analysis. Therefore it will be assumed that $p_0=0$, so that H_c can be neglected. An overbar will be used to indicate terms in this approximation. Then

$$\overline{H} = \overline{H}_a + \overline{H}_b. \tag{49}$$

A formula for $\overline{H}_a$ can be derived using Eqs. (15) and (A5b), with df expanded about r_0. Then with the aid of Eqs. (1), (44a), and (44b), one finds

$$\overline{H}_a = \frac{1}{2}\rho \int_0^{2\pi} \int_0^{\pi} \left[{}_1\Phi \frac{\partial_1\Phi}{\partial r} \right]_{r_0} r_0^2 \sin\theta \, d\theta \, d\varphi \, . \tag{50}$$

Now consider the formula for H_b,, given by Eq. (A7b). Expanding the integrand about r_0 yields

$$H_b = \alpha \int_0^{2\pi} \int_0^{\pi} \left\{ 1 + \frac{2\eta}{r_0} + \frac{1}{2r_0^2} \left[2\eta^2 + \left(\frac{\partial\eta}{\partial\theta} \right)^2 + \frac{1}{\sin^2\theta} \left(\frac{\partial\eta}{\partial\varphi} \right)^2 \right] + 0(\eta^3) \right\} r_0^2 \sin\theta \, d\theta \, d\varphi \, . \tag{51}$$

Next integrate the terms in square brackets in Eq. (51) by parts. Then use Eq. (44b) and also neglect terms $O(\eta^3)$. This yields the following formula for $\overline{H}_b$:

$$\overline{H}_b = \alpha \int_0^{2\pi} \int_0^{\pi} \left\{ r_0^2 + 2r_0 \, {}_1\eta + \frac{{}_1\eta}{2r_0^2} \left[2_1\eta - \left(\frac{\partial}{\partial\theta} \left(\sin\theta \frac{\partial_1\eta}{\partial\theta} \right) + \frac{1}{\sin^2\theta} \frac{\partial^2{}_1\eta}{\partial\varphi^2} \right) \right] \right\} \sin\theta \, d\theta \, d\varphi \, . \tag{52}$$

Equations (49), (50), and (52) will be used later in this paper to examine whether the energy is a constant of the motion at this level of perturbation theory.

V. SOLUTIONS OF PERTURBATION EQUATIONS

In this section solutions will be constructed for the first and second order systems of perturbation equations derived in Sec.IV. According to Eqs. (45a) and (45b), in every order of perturbation theory the velocity potential is required to satisfy Laplace's equation, Eq. (4), expressed in spherical coordinates and also satisfy a boundary condition at r=0 given by Eq. (5).

The velocity potential is time-dependent in general, and it can be expressed as a superposition of eigenfunctions of Laplace's equation with time dependent coefficients, as follows:

$$\Phi(r,\theta,\varphi;t)=\Phi_0(t)+\sum_{\ell,m,s} p^s_{\ell,m}(t)\,R_\ell(r)\,C^s_{\ell,m}(\theta,\varphi) \ . \tag{53a}$$

Laplace's equation and the boundary condition at r=0, Eq. (5), require that

$$R_\ell(r)=\left(\frac{r}{r_0}\right)^{\ell} . \tag{53b}$$

The coefficient $p^s_{\ell,m}(t)$ that appears in Eq. (3) can be written as a sum of perturbation terms, as follows:

$$p^s_{\ell,m}(t)={}_1p^s_{\ell,m}(t)+{}_2p^s_{\ell,m}(t)+\cdots . \tag{54}$$

Furthermore, the surface displacement can be expressed as

$$\eta(\theta,\varphi;t) = \sum_{\ell,m,s} z^s_{\ell,m}(t)\, C^s_{\ell,m}(\theta,\varphi)\,, \tag{55a}$$

where $z^s_{\ell,m}(t)$ can be written as a sum of perturbation terms, as follows:

$$z^s_{\ell,m}(t) = {}_1z^s_{\ell,m}(t) + {}_2z^s_{\ell,m}(t) + \cdots. \tag{55b}$$

A. Tesseral harmonics $C^s_{\ell,m}(\theta,\varphi)$

The functions $C^s_{\ell,m}(\theta,\varphi))$ that appear in Eqs. (53a) and (55a) are tesseral harmonics,[17] which provide a complete set for expanding functions of θ and φ. The tesseral harmonics are real valued functions that are essentially the $\cos(m\varphi)$ and $\sin(m\varphi)$ versions of the more familiar scalar spherical harmonics, $Y_{\ell,m}(\theta,\varphi)$. Properties of the $C^s_{\ell,m}(\theta,\varphi)$ that are important for this analysis will be summarized shortly. The ranges of summations in Eqs. (53) and (55) are as follows:

$$2 \le \ell \le \infty \quad 0 \le m \le \ell \quad s = 1 \text{ and } -1. \tag{56}$$

The $\ell = 1$ terms imply a shift of the center of mass of the drop from the original origin. This is demonstrated in Appendix B for an almost spherical drop. Therefore $\ell = 1$ must be excluded from the summation to enforce assumptions of the model stated earlier.

The functions $C^s_{\ell,m}(\theta,\varphi)$ for s=1 and s=-1 are given by the following formulas:

$$C^1_{\ell,m}(\theta,\varphi) = \frac{1}{\sqrt{2}}\left[Y_{\ell,m}(\theta,\varphi) + Y^*_{\ell,m}(\theta,\varphi)\right] \tag{57a}$$

$$C^{-1}_{\ell,m}(\theta,\varphi) = \frac{1}{i\sqrt{2}}\left[Y_{\ell,m}(\theta,\varphi) - Y^*_{\ell,m}(\theta,\varphi)\right], \tag{57b}$$

where

$$Y_{\ell,m}(\theta,\varphi) = Ne^{im\varphi}P_{\ell,m}(\theta) \tag{58a}$$

$$N = (-1)^m\left[\frac{(2\ell+1)}{4\pi}\frac{(\ell-m)!}{(\ell+m)!}\right]^{1/2} . \tag{58b}$$

The function $P_{\ell,m}(\theta)$ is an associated Legendre polynomial.

It can be shown that

$$Y^*_{\ell,m}(\theta,\varphi) = (-1)^m Y_{\ell,-m}(\theta,\varphi) . \tag{58c}$$

Note that Eqs. (57b) and (59) imply

$$C^{-1}_{\ell,m}(\theta,\varphi) = 0 \quad for \quad m = 0 . \tag{58d}$$

Also, the functions $P_{\ell,m}(\theta)$ satisfy

$$P_{\ell,-m}(\theta) = (-1)^m \frac{(\ell-m)!}{(\ell+m)!} P_{\ell,m}(\theta) . \tag{59a}$$

Equations (57a)-(59a) imply that the tesseral harmonics can be expressed as

$$C^{1}_{\ell,m}(\theta,\varphi) = \sqrt{2}N\,P_{\ell,m}(\theta)\cos m\varphi\,, \tag{59b}$$

$$C^{-1}_{\ell,m}(\theta,\varphi) = \sqrt{2}N\,P_{\ell,m}(\theta)\sin m\varphi\,. \tag{59c}$$

Spherical harmonics possess the following property:

$$\int_0^{2\pi}\int_0^{\pi} Y^{*}_{\ell,m}(\theta,\varphi)\,Y_{\ell',m'}(\theta,\varphi)\sin\theta\,d\theta\,d\varphi = \delta_{\ell,\ell'}\delta_{m,m'}\,. \tag{60}$$

With the aid of Eq. (60) one can show that the orthogonality and normalization relation for the tesseral harmonics is

$$\int_0^{2\pi}\int_0^{\pi} C^{s}_{\ell,m}(\theta,\varphi)\,C^{s'}_{\ell',m}(\theta,\varphi)\sin\theta\,d\theta\,d\varphi = N(\ell,m,s)\delta_{\ell,\ell'}\delta_{m,m'}\delta_{s,s'}\,, \tag{61}$$

where

$$For\ \ m \neq 0\ :\ N(\ell,m,s) = 1 \tag{62a}$$

$$For\ \ m = 0;\ s = 1\ :\ N(\ell,m,s) = 2 \tag{62b}$$

$$For\ \ m = 0;\ s = -1\ :\ N(\ell,m,s) = 0\,. \tag{62c}$$

The formulas for $C^{1}_{\ell,m}(\theta,\varphi)$ and $C^{-1}_{\ell,m}(\theta,\varphi)$ that appear in Eqs. (57a) and (57b) can be expressed as follows by using Eq. (58c):

$$C^{s}_{\ell,m}(\theta,\varphi) = \sum_{\lambda} a_m^{s,\lambda}\, Y_{\ell,(\lambda m)}(\theta,\varphi) \; . \tag{63}$$

Here the summation index λ takes on the values 1 and -1. The coefficients $a_m^{s,\lambda}$ are given by the following matrix equation:

$$\begin{pmatrix} a_m^{1,1} & a_m^{1,-1} \\ a_m^{-1,1} & a_m^{-1,-1} \end{pmatrix} = \begin{pmatrix} \frac{1}{\sqrt{2}} & (-1)^m \frac{1}{\sqrt{2}} \\ \frac{1}{i\sqrt{2}} & (-1)^m \frac{1}{i\sqrt{2}} \end{pmatrix}. \tag{64}$$

Using the property that the functions $C^{s}_{\ell,m}(\theta,\varphi)$ are real valued and also using Eqs. (60) and (63a), one can derive the following alternative form of Eq. (61):

$$\int_0^{2\pi}\int_0^{\pi} C^{s}_{\ell,m}(\theta,\varphi)\, C^{s'}_{\ell',m'}(\theta,\varphi) \sin\theta\, d\theta\, d\varphi = \delta_{\ell,\ell'} \sum_{\lambda,\lambda'} \left(a_m^{s,\lambda}\right)^* a_{m'}^{s',\lambda'}\, \delta_{(\lambda m),(\lambda' m')} \; . \tag{65}$$

All of these properties of tesseral harmonics will find useful application in what follows.

B. First order perturbation theory

The linearized equations based on Eqs. (45a) and (45b) with n=1 together with Eqs. (46a) and (47b) yield Rayleigh's results[12] for capillary waves in a liquid drop. This will be evident from what follows. The first step in solving these equations is to operate on Eq. (47b) with $\partial/\partial t$. This yields

$$\left({}_1\ddot{\Phi}\right)_{r_0} - \frac{\alpha}{\rho r_0^2}\left[2{}_1\dot{\eta} + \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial_1\dot{\eta}}{\partial\theta}\right) + \frac{1}{\sin^2\theta}\frac{\partial^2{}_1\dot{\eta}}{\partial\varphi^2}\right] = 0\,. \tag{66}$$

Substitution of Eq. (46a) into (66) gives, at $r=r_0$:

$${}_1\ddot{\Phi} - \frac{\alpha}{\rho r_0^2}\frac{\partial}{\partial r}\left[2{}_1\Phi + \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial_1\Phi}{\partial\theta}\right) + \frac{1}{\sin^2\theta}\frac{\partial^2{}_1\Phi}{\partial\varphi^2}\right] = 0\,. \tag{67}$$

Now substitute the expression

$${}_1\Phi(r,\theta,\varphi;t) = \sum_{\ell,m,s} {}_1p^s_{\ell,m}(t)R_\ell(r)C^s_{\ell,m}(\theta,\varphi)\,, \tag{68}$$

into Eq. (67). Then use Eqs. (57a) thru (58c) and also use the following equation satisfied by spherical harmonics:

$$\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{Y_{\ell,m}}{\partial\theta}\right) + \frac{1}{\sin^2\theta}\frac{\partial^2 Y_{\ell,m}}{\partial\varphi^2} = -\ell(\ell+1)Y_{\ell,m}\,. \tag{69}$$

One finds that at $r=r_0$,

$$\sum_{\ell,m,s}\left\{{}_1\ddot{p}^s{}_{\ell,m}(t)R_\ell(r_0) + \frac{\alpha}{\rho r_0^2}[\ell(\ell+1)-2]R'_\ell(r_0){}_1p^s_{\ell,m}(t)\right\}\, C^s_{\ell,m}(\theta,\varphi) = 0\,, \tag{70a}$$

where

$$R'_\ell(r) = \frac{dR_\ell(r)}{dr}\,. \tag{70b}$$

The tesseral harmonics are linearly independent; so Eq. (70a) implies that

$${}_1\ddot{p}_{\ell,m}(t) + \frac{\alpha}{\rho r_0^2}\left[\ell(\ell+1)-2\right]\frac{R'_\ell(r_0)}{R_\ell(r_0)}\,{}_1p^s_{\ell,m}(t) = 0\,. \tag{71a}$$

This equation for harmonic oscillation can be written as

$${}_1\ddot{p}^s_{\ell,m}(t) + \omega_\ell^2\;{}_1p^s_{\ell,m}(t) = 0\,, \tag{71b}$$

where

$$\omega_\ell^2 = \frac{\alpha}{\rho r_0^2}\left[\ell(\ell+1)-2\right]\frac{R'_\ell(r_0)}{R_\ell(r_0)}\,. \tag{71c}$$

With the aid of Eq. (53b) one finds

$$R'_\ell(r_0) = \frac{\ell}{r_0}\,. \tag{72}$$

Now the result in Eq. (71c) can be expressed as

$$\omega_\ell^2 = \frac{\alpha}{\rho r_0^3}\,\ell\left[\ell(\ell+1)-2\right]. \tag{73a}$$

which gives the frequency found by Rayleigh for capillary waves in an almost spherical liquid drop.

The general solution of Eq. (71c) is

$$ {}_1p^s_{\ell,m}(t) = {}_1p^s_{\ell,m}(0)\cos\left(\omega_\ell t + \alpha^s_{\ell,m}\right). \tag{74} $$

Next refer to Eqs. (55a) and (56) and write

$$ {}_1\eta(\theta,\varphi;t) = \sum_{\ell,m,s} {}_1z^s_{\ell,m}(t)C^s_{\ell,m}(\theta,\varphi). \tag{75} $$

Now substitute Eqs. (68) and (74) into Eq. (46a) and obtain

$$ \sum_{\ell,m,s} \left[{}_1\dot{z}^s_{\ell,m}(t) - {}_1p^s_{\ell,m}(t)R'_\ell(r_0)\right] C^s_{\ell,m}(\theta,\varphi) = 0. \tag{76a} $$

Linear independence of the tesseral harmonics implies that the individual coefficients in Eq. (76) vanish; therefore

$$ {}_1\dot{z}^s_{\ell,m}(t) - {}_1p^s_{\ell,m}(t)R'_\ell(r_0) = 0. \tag{76b} $$

Now operate on Eq. (76b) with $\int_0^t dt$ and take Eq. (74) into account. One finds

$$ {}_1 z^s_{\ell,m}(t) = \frac{R'_\ell(r_0)}{\omega_\ell}\, {}_1 p^s_{\ell,m}(0) \sin\left(\omega_\ell t + \alpha^s_{\ell,m}\right) \tag{77a} $$

if one takes ${}_1 z^s_{\ell,m}(0)$ to be given by

$$ {}_1 z^s_{\ell,m}(0) = \frac{R'_\ell(r_0)}{\omega_\ell}\, {}_1 p^s_{\ell,m}(0) \sin \alpha^s_{\ell,m} \,. \tag{77b} $$

In summary, the solutions of the first order equations of motion are given by

$$ {}_1\Phi(r,\theta,\varphi;t) = \sum_{\ell,m,s} {}_1 p^s_{\ell,m}(0) \cos\left(\omega_\ell t + \alpha^s_{\ell,m}\right) R_\ell(r_0) C^s_{\ell,m}(\theta,\varphi) \tag{78a} $$

$$ {}_1\eta(\theta,\varphi;t) = \sum_{\ell,m,s} \frac{1}{\omega_\ell} R'_\ell(r_0)\, {}_1 p^s_{\ell,m}(0) \sin\left(\omega_\ell t + \alpha^s_{\ell,m}\right) C^s_{\ell,m}(\theta,\varphi)\,, \tag{78b} $$

where ω_ℓ satisfies Eq. (71c).

C. Energy perturbation formula thru second order terms revisited

It is instructive to evaluate $\overline{H}$ in Eq. (49) using Eqs. (50), (52), (78a), and (78b). One finds

$$ \begin{aligned} \overline{H}_a = & \sum_{\ell,m,s} \sum_{\ell',m',s'} {}_1 p^s_{\ell,m}(0)\, {}_1 p^{s'}_{\ell',m'}(0) \cos\left(\omega_\ell t + \alpha^s_{\ell,m}\right) \cos\left(\omega_{\ell'} t + \alpha^{s'}_{\ell',m'}\right) \\ & \times r_0^2 R_\ell(r_0) R'_{\ell'}(r_0) \int_0^{2\pi} \int_0^{\pi} C^s_{\ell,m}(\theta,\varphi) C^{s'}_{\ell',m'}(\theta,\varphi) \sin\theta \, d\theta \, d\varphi \end{aligned} \tag{78c} $$

$$ = \frac{1}{2}\rho \sum_{\ell,m,s} N(\ell,m,s) r_0^2 R_\ell(r_0) R'_\ell(r_0)\, {}_1 p^s_{\ell,m}(0)\, {}_1 p^s_{\ell,m}(0) \cos^2\left(\omega_\ell t + \alpha^s_{\ell,m}\right) \tag{78d} $$

where Eq. (61) has been used to perform the integration over angles, and Kronecker deltas have been summed over.

Now consider evaluation of $\overline{H}_b$. Note that the term in Eq. (52) that is linear in ${}_1\eta$ vanishes when integrated over all angles. Substitution of Eq. (78a) into (52) and use of Eq. (69) yields

$$\begin{aligned}\overline{H}_b = 4\pi r_0^2\alpha + \frac{1}{2r_0^2}\sum_{\ell,m,s}\sum_{\ell',m',s'}\Bigg\{&[2+\ell'(\ell'+1)]\\ &\times\frac{1}{\omega_\ell\omega_{\ell'}}R_\ell'(r_0)R_{\ell'}'(r_0)\;{}_1p_{\ell,m}^s(0)\;{}_1p_{\ell',m'}^{s'}(0)\sin\left(\omega_\ell t+\alpha_{\ell,m}^s\right)\sin\left(\omega_{\ell'}t+\alpha_{\ell',m'}^{s'}\right)\\ &\times\int_0^{2\pi}\int_0^{\pi} C_{\ell,m}^s(\theta,\varphi)C_{\ell',m'}^{s'}(\theta,\varphi)\sin\theta\,d\theta\,d\varphi\Bigg\}.\end{aligned} \tag{78e}$$

Next use Eq. (61) and then sum over Kronecker deltas. The result is

$$\begin{aligned}\overline{H}_b = \Bigg\{&4\pi r_0^2\alpha + \frac{1}{2r_0^2}\sum_{\ell,m,s} N(\ell,m,s)\left([\ell(\ell+1)-2]+4\right)\\ &\times\frac{1}{\omega_\ell^2}R_\ell'(r_0)R_\ell'(r_0)\;{}_1p_{\ell,m}^s(0)\;{}_1p_{\ell,m}^s(0)\sin^2\left(\omega_\ell t+\alpha_{\ell,m}^s\right)\Bigg\}.\end{aligned} \tag{78f}$$

Substitute the expression for ω_ℓ^2 in Eq. (71c) into Eq. (78f), and obtain

$$\begin{aligned}\overline{H}_b = \Bigg\{&4\pi r_0^2\alpha + \frac{1}{2}\rho\sum_{\ell,m,s} N(\ell,m,s)R_\ell(r_0)R_\ell'(r_0)\;{}_1p_{\ell,m}^s(0)\;{}_1p_{\ell,m}^s(0)\sin^2\left(\omega_\ell t+\alpha_{\ell,m}^s\right)\\ &+\frac{1}{2}\rho\sum_{\ell,m,s}\frac{4}{[\ell(\ell+1)-2]}N(\ell,m,s)R_\ell(r_0)R_\ell'(r_0)\;{}_1p_{\ell,m}^s(0)\;{}_1p_{\ell,m}^s(0)\sin^2\left(\omega_\ell t+\alpha_{\ell,m}^s\right)\Bigg\}.\end{aligned} \tag{78g}$$

Next combine Eqs. (78d), (78g), and (49) and then use the relation $\sin^2 x + \cos^2 x = 1$. One finds

$$\bar{H} = \left\{ 4\pi r_0^2 \alpha + \frac{1}{2}\rho \sum_{\ell,m,s} N(\ell,m,s) R_\ell(r_0) R'_\ell(r_0)\ {}_1p^s_{\ell,m}(0)\ {}_1p^s_{\ell,m}(0) \right.$$
$$\left. + \frac{1}{2}\rho \sum_{\ell,m,s} \frac{4}{[\ell(\ell+1)-2]} N(\ell,m,s) R_\ell(r_0) R'_\ell(r_0)\ {}_1p^s_{\ell,m}(0)\ {}_1p^s_{\ell,m}(0) \sin^2\left(\omega_\ell t + \alpha^S_{\ell,m}\right) \right\}. \tag{78h}$$

The quantity in square brackets in Eq. (78h) is a constant of the motion. However, the last term in Eq. (78h) shows that at this level of approximation the energy has a time dependence.

D. Second order perturbation theory

The second order theory based on Eqs. (46b) and (47c) includes the leading nonlinear interactions among capillary waves when the waves are defined by the first order theory.

The first step in solving these equations is to operate on Eq. (47c) with $\partial / \partial t$. This yields

$$\left\{ \left({}_2\ddot{\Phi}\right)_{r_0} + \left(\frac{\partial}{\partial r}{}_1\ddot{\Phi}\right)_{r_0} {}_1\eta + \left(\frac{\partial}{\partial r}{}_1\dot{\Phi}\right)_{r_0} {}_1\dot{\eta} + \left[\left(\vec{\nabla}_1\Phi\right)\cdot\left(\vec{\nabla}_1\dot{\Phi}\right)\right]_{r_0} \right.$$

$$- \frac{\alpha}{\rho r_0^2}\left[2\,{}_2\dot{\eta} + \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial\,{}_2\dot{\eta}}{\partial\theta}\right) + \frac{1}{\sin^2\theta}\frac{\partial^2}{\partial\varphi^2}{}_2\dot{\eta} \right.$$

$$- \frac{2}{r_0}{}_1\dot{\eta}\left({}_1\eta + \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial}{\partial\theta}{}_1\eta\right) + \frac{1}{\sin^2\theta}\frac{\partial^2}{\partial\varphi^2}{}_1\eta \right) \tag{79}$$

$$\left.\left. - \frac{2}{r_0}{}_1\eta\left({}_1\dot{\eta} + \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial}{\partial\theta}{}_1\dot{\eta}\right) + \frac{1}{\sin^2\theta}\frac{\partial^2}{\partial\varphi^2}{}_1\dot{\eta} \right)\right]\right\} = 0 .$$

Eq. (79) can be converted to the following form with the aid of Eqs. (46a) and (46b). At $r=r_0$,

$${}_2\ddot{\Phi} - \frac{\alpha}{\rho r_0^2}\frac{\partial}{\partial r}\left[2\,{}_2\Phi + \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial}{\partial\theta}{}_2\Phi\right) + \frac{1}{\sin^2\theta}\frac{\partial^2}{\partial\varphi^2}{}_2\Phi \right] = P(r_0,\theta,\varphi;t) . \tag{80}$$

The linear operator that acts on ${}_2\Phi$ on the left hand side of this equation is the same operator that acts on ${}_1\Phi$ in Eq. (67). The right hand side of Eq. (80) does not involve ${}_2\Phi$. However, it contains terms that are second order in ${}_1\Phi$ and ${}_1\eta$, as indicated next, where all terms are evaluated at $r=r_0$:

$$P(r_0,\theta,\varphi;t) = \sum_{1\le j\le 7} P_j(r_0,\theta,\varphi;t) \tag{81a}$$

$$P_1 = \frac{\alpha}{\rho r_0^2}\left[2 + \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial}{\partial\theta}\right) + \frac{1}{\sin^2\theta}\frac{\partial^2}{\partial\varphi^2} \right]\left[\left(\frac{\partial^2}{\partial r^2}{}_1\Phi\right){}_1\eta\right] \tag{81b}$$

$$P_2 = -{}_1\eta\frac{\partial}{\partial r}{}_1\ddot{\Phi} \tag{81c}$$

$$P_3 = -{}_1\dot{\eta}\frac{\partial}{\partial r}{}_1\dot{\Phi} \tag{81d}$$

$$P_4 = -\frac{2\alpha}{\rho r_0^3}\left\{\left[2+\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial}{\partial\theta}\right)+\frac{1}{\sin^2\theta}\frac{\partial^2}{\partial\varphi^2}\right]{}_1\eta\right\}{}_1\dot{\eta} \tag{81e}$$

$$P_5 = -\frac{2\alpha}{\rho r_0^3}\,{}_1\eta\left[\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial}{\partial\theta}\right)+\frac{1}{\sin^2\theta}\frac{\partial^2}{\partial\varphi^2}\right]{}_1\dot{\eta} \tag{81f}$$

$$P_6 = -\left(\vec{\nabla}_1\dot{\Phi}\right)\cdot\left(\vec{\nabla}_1\Phi\right) \tag{81g}$$

$$P_7 = -\frac{\alpha}{\rho r_0^2}\left[2+\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial}{\partial\theta}\right)+\frac{1}{\sin^2\theta}\frac{\partial^2}{\partial\varphi^2}\right]\left[\left(\vec{\nabla}_1\eta\right)\cdot\left(\vec{\nabla}_1\Phi\right)\right]. \tag{81h}$$

E. Evaluation of terms in P involving only scalar functions

The functions P_1 thru P_5 depend only on scalar quantities and they can be readily evaluated using Eqs. (63) and (69) together the completeness property of scalar spherical harmonics. The completeness property allows one to write

$$Y_{L_2,M_2}(\theta,\varphi)Y_{L_1,M_1}(\theta,\varphi) = \sum_{L_3,M_3} b_{L_3,M_3}(L_1,M_1;L_2,M_2)Y_{L_3,M_3}(\theta,\varphi)\ . \tag{82}$$

The coefficients b_{L_3,M_3} can be evaluated using the relation

$$\int_0^{2\pi}\int_0^{\pi} Y^*_{L_3,M_3}(\theta,\varphi)Y_{L_2,M_2}(\theta,\varphi)Y_{L_1,M_1}(\theta,\varphi)\sin\theta\,d\theta\,d\varphi$$
$$= \left[\frac{(2L_1+1)(2L_2+1)}{4\pi(2L_3+1)}\right]^{1/2} C(L_1,L_2,L_3;M_1,M_2)C(L_1,L_2,L_3;0,0)\ . \tag{83}$$
$$\times\ \delta_{M_3,(M_1+M_2)}\ .$$

Here an abbreviated notation for Clebsch-Gordan (C-G) coefficients $C(L_1,L_2,L_3;M_1,M_2)$ adopted by Rose[18] has been used. A third M index is not expressed explicitly in this notation, but it is understood that $M_3=M_1+M_2$ in any C-G coefficient that does not vanish. The factor $\delta_{M_3,(M_1+M_2)}$ is therefore redundant, but it is retained here for clarity. With the aid of Eqs. (60), (82), and (83), one finds

$$\begin{aligned} & b_{L_3,M_3}(L_1,M_1;L_2,M_2) \\ & \quad = \left[\frac{(2L_1+1)(2L_2+1)}{4\pi(2L_3+1)}\right]^{1/2} C(L_1,L_2,L_3;M_1,M_2)\,C(L_1,L_2,L_3;0,0) \; . \\ & \quad \times \delta_{M_3,(M_1+M_2)} \; . \end{aligned} \tag{84}$$

To evaluate P_1, first refer to Eqs. (63), (68), and (75) and write

$$_1\Phi(r,\theta,\varphi;t) = \sum_{\ell,m,s,\lambda} {}_1p^s_{\ell,m}(t)\; R_\ell(r)\; a_m^{s,\lambda}\; Y_{\ell,(\lambda m)}(\theta,\varphi) \tag{85}$$

$$_1\eta(\theta,\varphi;t) = \sum_{\ell,m,s,\lambda} {}_1z^s_{\ell,m}(t)\; a_m^{s,\lambda}\; Y_{\ell,(\lambda m)}(\theta,\varphi)\,. \tag{86}$$

Recall that $_1\eta$ is real valued, so that $_1\eta={}_1\eta^*$. Furthermore, $z^s_{\ell,m}(t) = \left(z^s_{\ell,m}(t)\right)^*$ since $C_{\ell,m}(\theta,\varphi)$ is real valued. The operand in Eq. (81b) can then be treated as follows:

$$
{}_1\eta\left(\frac{\partial^2}{\partial r^2}{}_1\Phi\right)_{r_0} = {}_1\eta^*\left(\frac{\partial^2}{\partial r^2}{}_1\Phi\right)_{r_0} \tag{87a}
$$

$$
= \sum_{\ell,m,s,\lambda}\sum_{\ell',m',s',\lambda'}\left[{}_1 z^s_{\ell,m}(t)\, a^{s,\lambda}_m\, Y^s_{\ell,(\lambda m)}(\theta,\varphi)\right]^*\left[{}_1 p^{s'}_{\ell',m'}(t) R''_{\ell'}(r_0) a^{s',\lambda'}_{m'}\, Y_{\ell',(\lambda' m')}(\theta,\varphi)\right] \tag{87b}
$$

$$
= \sum_{\ell,m,s,\lambda}\;\sum_{\ell',m',s',\lambda'}\left(a^{s,\lambda}_m\right)^* a^{s',\lambda'}_{m'} R''_{\ell'}(r_0)\; {}_1 z^s_{\ell,m}(t)\; {}_1 p^{s'}_{\ell',m'}(t)(-1)^{(\lambda m)}
$$
$$
\times\; Y_{\ell,(-\lambda m)}(\theta,\varphi) Y_{\ell',(\lambda' m')}(\theta,\varphi)\,, \tag{87c}
$$

where Eq. (58c) has been used in the last step.

Next substitute Eq. (87c) into (81b). and rewrite the result with the aid of Eqs.(82) and (69). The following formula for P_1 then emerges:

$$
P_1 = \sum_{L_1,M_1}\;\sum_{\ell,m,s,\lambda}\;\sum_{\ell',m',s',\lambda'}\left(a^{s,\lambda}_m\right)^* a^{s',\lambda'}_{m'}\; {}_1 D_{L_1,M_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0)
$$
$$
\times\; {}_1 z^s_{\ell,m}(t)\; {}_1 p^{s'}_{\ell',m'}(t) Y_{L_1,M_1}(\theta,\varphi)\,, \tag{88}
$$

where

$$
{}_1 D_{L_1,M_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0)
$$
$$
= \frac{\alpha}{\rho {r_0}^2}(-1)^{(\lambda m)}\left[2 - L_1(L_1+1)\right] R''_{\ell'}(r_0)\, b_{L_1,M_1}(\ell,(\lambda m);\ell',(\lambda' m'))\,. \tag{89}
$$

The function P_2 in Eq. (81c) will be treated next.. First use Eqs. (71b) and (85) and obtain

$$
\left(\frac{\partial}{\partial r}\,{}_1\Phi\right)_{r_0} = \sum_{\ell,m,s,\lambda}(-1)\omega^2_\ell\;\; {}_1 p^s_{\ell,m}(t)\, R'_\ell(r_0)\, Y_{\ell,(\lambda m)}(\theta,\varphi) \tag{90}
$$

Then use a procedure similar to that for P_1, and find

$$P_2 = \sum_{L_1,M_1} \sum_{\ell,m,s,\lambda} \sum_{\ell',m',s',\lambda'} \left(a_m^{s,\lambda}\right)^* a_{m'}^{s',\lambda'} {}_2D_{L_1,M_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0) \times {}_1z_{\ell,m}^{s}(t)\, {}_1p_{\ell',m'}^{s'}(t) Y_{L_1,M_1}(\theta,\varphi) , \tag{91}$$

where

$${}_2D_{L_1,M_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0) = (-1)^{(\lambda m)} R'_{\ell'}(r_0)\omega_{\ell'}^2\; b_{L_1,M_1}(\ell,(\lambda m);\ell',(\lambda' m')) . \tag{92}$$

Turn next to evaluation of P_3 in Eq. (81d). Note that

$${}_1\dot{\eta} = -\left(\frac{\partial}{\partial r}{}_1\Phi\right)_{r_0} . \tag{93a}$$

Then use Eq. (85) and obtain

$$\left(\frac{\partial}{\partial r}{}_1\Phi\right)_{r_0} = \sum_{\ell,m,s,\lambda} {}_1p_{\ell,m}^{s}(t) R'_\ell(r_0) a_m^{s,\lambda} Y_{\ell,(\lambda m)}(\theta,\varphi) . \tag{93b}$$

Take the time derivative of Eq. (93b) and obtain

$$\left(\frac{\partial}{\partial r}{}_1\dot{\Phi}\right)_{r_0} = \sum_{\ell,m,s,\lambda} {}_1\dot{p}_{\ell,m}^{s}(t) R'_\ell(r_0) a_m^{s,\lambda} Y_{\ell,(\lambda m)}(\theta,\varphi) . \tag{94}$$

Then with the aid of Eqs. (74) and (77a), one finds

$$
{}_1\dot{p}^{s}_{\ell,m}(t) = -\frac{\omega_\ell^2}{R'_\ell(r_0)}\,{}_1z^{s}_{\ell,m}(t)\,. \tag{95}
$$

Combine Eqs. (93a) thru (95) with (81d) and then apply the same procedure as before. The result is

$$
\begin{aligned}
P_3 = \sum_{L_1,M_1}\ \sum_{\ell,m,s,\lambda}\ \sum_{\ell',m',s',\lambda'} \left(a_m^{s,\lambda}\right)^* a_{m'}^{s',\lambda'}\,{}_3D_{L_1,M_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0) \\
\times\ {}_1z^{s}_{\ell,m}(t)\ {}_1p^{s'}_{\ell',m'}(t) Y_{L_1,M_1}(\theta,\varphi)\,,
\end{aligned} \tag{96}
$$

where

$$
{}_3D_{L_1,M_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0) = (-1)^{(\lambda m)} R'_{\ell'}(r_0)\omega_\ell^2 b_{L_1,M_1}(\ell,(\lambda m);\ell',(\lambda' m'))\,. \tag{97}
$$

Next evaluate the function P_4 in Eq. (81e). Use Eqs. (86), (93a), (93b), and (69). Then apply the same procedure as before. One finds

$$
\begin{aligned}
P_4 = \sum_{L_1,M_1}\ \sum_{\ell,m,s,\lambda}\ \sum_{\ell',m',s',\lambda'} \left(a_m^{s,\lambda}\right)^* a_{m'}^{s',\lambda'}\,{}_4D_{L_1,M_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0) \\
\times\ {}_1z^{s}_{\ell,m}(t)\ {}_1p^{s'}_{\ell',m'}(t) Y_{L_1,M_1}(\theta,\varphi)\,,
\end{aligned} \tag{98}
$$

where

$$
\begin{aligned}
&{}_4D_{L_1,M_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0) \\
&= (-1)^{[1+(\lambda m)]}\frac{2\alpha}{\rho r_0^3}[2-\ell(\ell+1)]R'_{\ell'}(r_0) b_{L_1,M_1}(\ell,(\lambda m);\ell',(\lambda' m'))\,.
\end{aligned} \tag{99}
$$

The function P_5 in Eq. (81f) can be evaluated as follows. First use Eqs. (93a), (93b), and (69) to simplify the factor involving ${}_1\dot{\eta}$. Then use Eq. (86) to rewrite the factor ${}_1\eta$. Substitute these results into Eq. (81f) and apply Eq. (82) to the subsequent equation. One finds

$$P_5 = \sum_{L_1,M_1} \sum_{\ell,m,s,\lambda} \sum_{\ell',m',S',\lambda'} \left(a_m^{s,\lambda}\right)^* a_{m'}^{s',\lambda'} \, {}_5D_{L_1,M_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0) \times {}_1z_{\ell,m}^{s}(t) \, {}_1p_{\ell',m'}^{s'}(t) Y_{L_1,M_1}(\theta,\varphi) \,, \tag{100}$$

where

$${}_5D_{L_1,m_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0) = \frac{2\alpha}{\rho r_0^3}(-1)^{(\lambda m)}\left[\ell'(\ell'+1)\right]R_{\ell'}'(r_0) b_{L_1,M_1}\left(\ell,(\lambda m);\ell',(\lambda' m')\right). \tag{101}$$

This completes evaluation of terms involving only scalar functions.

F. Evaluation of terms involving vector functions

Turn next to evaluation of P_6 and P_7, which involve dot products of two vectors. The gradient formula,[19] which occurs in treatments of angular momentum in quantum mechanics, is the key to evaluation of these terms. The gradient formula is as follows:

$$\vec{\nabla}\left[\psi(r)Y_{L,M}(\theta,\varphi)\right]$$
$$=\left\{-\left(\frac{\ell+1}{2\ell+1}\right)^{1/2}\left(\frac{d}{dr}-\frac{\ell}{r}\right)\psi(r)\vec{Y}^{m}_{\ell,\ell+1}(\theta,\varphi)+\left(\frac{\ell}{2\ell+1}\right)^{1/2}\left(\frac{d}{dr}-\frac{\ell+1}{r}\right)\psi(r)\vec{Y}^{m}_{\ell,\ell-1}(\theta,\varphi)\right. \tag{102}$$

Here $\psi(r)$ may be any differentiable function of r, $Y_{\ell,m}(\theta,\varphi)$ is a scalar spherical harmonic, and $\vec{Y}^{m}_{\ell,\ell+1}(\theta,\varphi)$ and $\vec{Y}^{m}_{\ell,\ell-1}(\theta,\varphi)$ are vector spherical harmonics.[19,20] Some properties of vector spherical harmonics that are important for this analysis are given in Appendix C, and the following result is established there:

$$\left(\vec{Y}^{M}_{J,\ell}(\theta,\varphi)\right)^{*}\cdot\vec{Y}^{M'}_{J',\ell'}(\theta,\varphi)=\sum_{L_1,M_1} a_{L_1,M_1}(J,\ell,M;J',\ell',M')Y_{L_1,M_1}(\theta,\varphi). \tag{103}$$

A formula for a_{L_1,M_1} is also derived in Appendix C.

With the aid of Eq. (85) and the time derivative of that equation, one can write P_6 in Eq. (81g) as follows:

$$P_6=\sum_{\ell,m,s,\lambda}\ \sum_{\ell',m',s',\lambda'}(-1)\left[{}_1\dot{p}_{\ell,m}(t)a^{s,\lambda}_{m}\right]^{*}{}_1p^{s'}_{\ell',m'}(t)a^{s',\lambda'}_{m'}$$
$$\times\left\{\left[\vec{\nabla}\left(R_\ell(r)Y_{\ell,(\lambda m)}(\theta,\varphi)\right)\right]^{*}\cdot\left[\vec{\nabla}\left(R_{\ell'}(r)Y_{\ell',(\lambda' m')}(\theta,\varphi)\right)\right]\right\}_{r_0} \tag{104}$$

Next apply Eq. (102) to rewrite the gradient terms in Eq. (104), and also use Eq. (95) to express ${}_1\dot{p}^{s}_{\ell,m}(t)$ in terms of ${}_1z^{s}_{\ell,m}(t)$. Then apply Eq. (103) to the result. One finds

$$P_6 = \sum_{L_1,M_1} \sum_{\ell,m,s,\lambda} \sum_{\ell',m',s',\lambda'} \left(a_m^{s,\lambda}\right)^* a_{m'}^{s',\lambda'} \, {}_6D_{L_1,M_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0) \times {}_1z_{\ell,m}^{s}(t) \, {}_1p_{\ell',m'}^{s'}(t) Y_{L_1,M_1}(\theta,\varphi) , \tag{105}$$

where

$$\begin{aligned}
&{}_6D_{L_1,M_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0) \\
&= \left(\frac{\omega_\ell^2}{R_\ell'(r_0)}\right) \\
&\times\Bigg\{\left(\frac{\ell+1}{2\ell+1}\right)^{1/2}\left(R_\ell'(r_0)-\frac{\ell}{r_0}R_\ell(r_0)\right)\left(\frac{\ell'+1}{2\ell'+1}\right)^{1/2}\left(R_{\ell'}'(r_0)-\frac{\ell'}{r_0}R_{\ell'}(r_0)\right)a_{L_1,M_1}(\ell,\ell+1,(\lambda m);\ell',\ell'+1,(\lambda' m')) \\
&-\left(\frac{\ell+1}{2\ell+1}\right)^{1/2}\left(R_\ell'(r_0)-\frac{\ell}{r_0}R_\ell(r_0)\right)\left(\frac{\ell'+1}{2\ell'+1}\right)^{1/2}\left(R_{\ell'}'(r_0)+\frac{\ell'+1}{r_0}R_{\ell'}(r_0)\right)a_{L_1,M_1}(\ell,\ell+1,(\lambda m);\ell',\ell'-1,(\lambda' m')) \\
&-\left(\frac{\ell}{2\ell+1}\right)^{1/2}\left(R_\ell'(r_0)+\frac{\ell+1}{r_0}R_\ell(r_0)\right)\left(\frac{\ell'+1}{2\ell'+1}\right)^{1/2}\left(R_{\ell'}'(r_0)-\frac{\ell'}{r_0}R_{\ell'}(r_0)\right)a_{L_1,M_1}(\ell,\ell-1,(\lambda m);\ell',\ell'+1,(\lambda' m')) \\
&+\left(\frac{\ell}{2\ell+1}\right)^{1/2}\left(R_\ell'(r_0)+\frac{\ell+1}{r_0}R_\ell(r_0)\right)\left(\frac{\ell'}{2\ell'+1}\right)^{1/2}\left(R_{\ell'}'(r_0)+\frac{\ell'+1}{r_0}R_{\ell'}(r_0)\right)a_{L_1,M_1}(\ell,\ell-1,(\lambda m);\ell',\ell'-1,(\lambda' m'))\Bigg\} .
\end{aligned} \tag{106}$$

The term P_7 in Eq. (81h) can also be evaluated with the aid of the gradient formula. The quantity $[(\nabla_1\eta)\bullet(\nabla_1\Phi)]$ can be rewritten with the aid of Eqs. (85), (86), (102), and (103). Applying Eq. (69) to that result, one can complete the evaluation of P_7. The result is

$$P_7 = \sum_{L_1,M_1} \sum_{\ell,m,s,\lambda} \sum_{\ell',m',s',\lambda'} \left(a_m^{s,\lambda}\right)^* a_{m'}^{s',\lambda'} \, {}_7D_{L_1,M_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0)$$
$$\times \, {}_1z_{\ell,m}^{s}(t) \, {}_1p_{\ell',m'}^{s'}(t) Y_{L_1,M_1}(\theta,\varphi), \tag{107}$$

where

$${}_7D_{L_1,M_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0)$$
$$= -\frac{\alpha}{\rho r_0^2}\left[2 - L_1(L_1+1)\right]$$
$$\times\left\{\left(\frac{\ell+1}{2\ell+1}\right)^{1/2}\left(-\frac{\ell}{r_0}\right)\left(\frac{\ell'+1}{2\ell'+1}\right)\left(R'_{\ell'}(r_0) - \frac{\ell'}{r_0}R_{\ell'}(r_0)\right)a_{L_1,M_1}(\ell,\ell+1,(\lambda m);\ell',\ell'+1,(\lambda' m'))\right.$$
$$-\left(\frac{\ell+1}{2\ell+1}\right)^{1/2}\left(-\frac{\ell}{r_0}\right)\left(\frac{\ell'}{2\ell'+1}\right)^{1/2}\left(R'_{\ell'}(r_0) + \frac{\ell'+1}{r_0}R_{\ell'}(r_0)\right)a_{L_1,M_1}(\ell,\ell+1,(\lambda m);\ell',\ell'-1,(\lambda' m')) \tag{108}$$
$$-\left(\frac{\ell}{2\ell+1}\right)^{1/2}\left(\frac{\ell+1}{r_0}\right)\left(\frac{\ell'+1}{2\ell'+1}\right)^{1/2}\left(R'_{\ell'}(r_0) - \frac{\ell'}{r_0}R_{\ell'}(r_0)\right)a_{L_1,M_1}(\ell,\ell-1,(\lambda m);\ell',\ell'+1,(\lambda' m'))$$
$$\left.+\left(\frac{\ell}{2\ell+1}\right)^{1/2}\left(\frac{\ell+1}{r_0}\right)\left(\frac{\ell'}{2\ell'+1}\right)^{1/2}\left(R'_{\ell'}(r_0) + \frac{\ell'+1}{r_0}R_{\ell'}(r_0)\right)a_{L_1,M_1}(\ell,\ell-1,(\lambda m);\ell',\ell'+1,(\lambda' m'))\right\}.$$

G. Derivation of differential equation satisfied by ${}_2p_{L,M}^{S}(t)$

The differential equation satisfied by the coefficients ${}_2p_{L,M}^{S}(t)$ will be considered next. Equations (53a)-(56) imply that ${}_2\Phi$ and ${}_2\eta$ may be represented as

$${}_2\Phi(r,\theta,\varphi;t) = \sum_{\ell,m,s} {}_2p_{\ell,m}^{s}(t) R_\ell(r) C_{\ell,m}^{s}(\theta,\varphi) \tag{109}$$

$${}_2\eta(\theta,\varphi;t) = \sum_{\ell,m,s} {}_2z_{\ell,m}^{s}(t) C_{\ell,m}^{s}(\theta,\varphi). \tag{110}$$

Substitution of Eq. (109) into (80) and use of Eqs. (69) and (71c) yields

$$\sum_{L_1,M_1,S_1}\left[{}_2\ddot{p}^{S_1}_{L_1,M_1}(t)+\omega^2_{L_1}\ {}_2p^{S_1}_{L_1,M_1}(t)\right]R_{L_1}(r_0)\ C^{S_1}_{L_1,M_1}(\theta,\varphi)=P(r_0,\theta,\varphi;t) \tag{111}$$

Now multiply Eq. (111) by $C^{S}_{L,M}(\theta,\varphi)$ and integrate over the unit sphere. Use Eq. (61) to evaluate the result of this operation on the left hand side of Eq. (111). Use Eqs. (60) and (63) and the fact that the tesseral harmonics are real valued to evaluate the result of this operation on the right hand side of Eq. (111). Then one obtains

$$\begin{aligned}
&\sum_{L_1,M_1,S_1}\left[{}_2\ddot{p}^{S_1}_{L_1,M_1}(t)+\omega^2_{L_1}\ {}_2p^{S_1}_{L_1,M_1}(t)\right]R_{L_1}(r_0)\ N(L_1,M_1,S_1)\delta_{L,L_1}\delta_{M,M_1}\delta_{S,S_1}\\
&=\sum_{j,\Lambda,L_1,M_1}\ \sum_{\ell,m,s,\lambda}\ \sum_{\ell',m',s',\lambda'}\left(a^{S,\Lambda}_{M}\right)^*\left(a^{s,\lambda}_{m}\right)^* a^{s',\lambda'}_{m'}\\
&\times\ {}_jD_{L_1,M_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0)\ {}_1z^{s}_{\ell,m}(t)\ {}_1p^{s'}_{\ell',m'}(t)\delta_{L,L_1}\delta_{(\Lambda M),M_1}\ .
\end{aligned} \tag{112}$$

where the range for j is $1\le j\le 7$.

Refer to Eqs. (59c) and (62a)-(62c) and note that the only cases in which $N(\ell,m,s)$ vanishes are those where $C^{-1}_{\ell,m}(\theta,\varphi)$ itself vanishes. Therefore those terms give no contribution to ${}_2\Phi$ and ${}_2\eta$. In what follows we will specify that these terms are not included in the summations. Then $N(\ell,m,s)\neq 0$ in the terms that actually occur in Eq. (112). Now perform the sum over Kronecker deltas in Eq. (112). Then use Eqs. (74) and (77a) in the right hand side of that equation. Simple algebraic rearrangement of the subsequent equation yields the following result:

$$
\begin{aligned}
&{}_2\ddot{p}^{S}_{L,M}(t) + \omega_L^2 \, {}_2p^{S}_{L,M}(t) \\
&= \sum_{\Lambda} \sum_{\ell,m,s,\lambda} \sum_{\ell',m',s',\lambda'} E_{L,M,S,\Lambda}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0) \\
&\times {}_1p^{s}_{\ell,m}(0) \, {}_1p^{s'}_{\ell',m'}(0) \, \sin\left(\omega_\ell t + \alpha^{s}_{\ell,m}\right)\cos\left(\omega_{\ell} t + \alpha^{s'}_{\ell',m'}\right),
\end{aligned} \tag{113}
$$

where

$$
\begin{aligned}
&E_{L,M,S,\Lambda}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0) \\
&= \left[\frac{R'_\ell(r_0)}{N(L,M,S)R_L(r_0)\omega_\ell}\right]\left(a^{S,\Lambda}_{M}\right)^{*}\left(a^{s,\lambda}_{m}\right)^{*} a^{s',\lambda'}_{m'} \\
&\times \sum_{1\le j\le 7} {}_jD_{L,(\Lambda M)}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0).
\end{aligned} \tag{114}
$$

Equation (113) is the relation that we wished to derive.

H. Time dependence of second order coefficients

Solving Eq. (113) for the time dependent amplitudes ${}_2p^{S}_{L,M}(t)$ is the task that will be addressed next. Toward that end, we shall first express the time dependent sines and cosines in Eq. (113) in terms of complex exponentials. The result is

$$
\begin{aligned}
&{}_2\ddot{p}^S_{L,M}(t) + \omega_L^2\ {}_2p^S_{L,M}(t) \\
&= \sum_{\Lambda} \sum_{\ell,m,s,\lambda} \sum_{\ell',m',s',\lambda'} E_{L,M,S,\Lambda}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0)\ {}_1p^s_{\ell,m}(0)\ {}_1p^{s'}_{\ell',m'}(0) \\
&\times \frac{1}{2i}\Big\{\exp\Big(i\Big[(\omega_\ell + \omega_{\ell'})t + \big(\alpha^s_{\ell,m} + \alpha^{s'}_{\ell',m'}\big)\Big]\Big) \\
&- \exp\Big(-i\Big[(\omega_\ell + \omega_{\ell'})t + \big(\alpha^s_{\ell,m} + \alpha^{s'}_{\ell',m'}\big)\Big]\Big) \\
&+ \exp\Big(i\Big[(\omega_\ell - \omega_{\ell'})t + \big(\alpha^s_{\ell,m} - \alpha^{s'}_{\ell',m'}\big)\Big]\Big) \\
&- \exp\Big(-i\Big[(\omega_\ell - \omega_{\ell'})t + \big(\alpha^s_{\ell,m} - \alpha^{s'}_{\ell',m'}\big)\Big]\Big)\Big\}.
\end{aligned}
\tag{115}
$$

The complete solution of Eq. (115) can be constructed as a linear combination of solutions of a differential equation of the following form:

$$\ddot{\psi} + \omega^2\psi = e^{i\omega' t}. \tag{116}$$

The solution of Eq.(116) is[7]:

$$\psi(t) = \frac{e^{i\omega' t}}{\omega^2 - (\omega')^2} - \frac{1}{2\omega}\left(\frac{e^{i\omega t}}{\omega - \omega'} + \frac{e^{-i\omega t}}{\omega + \omega'}\right) \quad for\ \omega^2 \neq (\omega')^2 \tag{117a}$$

$$\psi(t) = \frac{te^{i\omega' t}}{2i\omega'} + \frac{1}{4\omega\omega'}\left(e^{i\omega t} - e^{-i\omega t}\right) \quad for\ \omega^2 = (\omega')^2 . \tag{117b}$$

The steps involved in obtaining the solution are shown in Appendix D.

In order to express the solution of Eq. (113) efficiently, it is useful to first introduce functions ${}_1S_L$, ${}_2S_L$, ${}_3S_L$, ${}_4S_L$. It is specified that ω_L, ω_ℓ, and $\omega_{\ell'}$ are all different from zero. This takes into account that L, ℓ, and ℓ' are different from one. Then

For $\omega_L^2 \neq (\omega_\ell + \omega_{\ell'})^2$:

$$
\begin{aligned}
&{}_1S_L(\ell,m,s;\ell',m',s';t) \\
&= \left\{ \frac{\sin\left[(\omega_\ell + \omega_{\ell'})t + \left(\alpha_{\ell,m}^s + \alpha_{\ell',m'}^{s'}\right)\right]}{\omega_L^2 - (\omega_\ell + \omega_{\ell'})^2} \right. \qquad (118) \\
&\left. - \frac{1}{2\omega_L} \left(\frac{\sin\left[\omega_L t + \left(\alpha_{\ell,m}^s + \alpha_{\ell',m'}^{s'}\right)\right]}{\omega_L - (\omega_\ell + \omega_{\ell'})} - \frac{\sin\left[\omega_L t - \left(\alpha_{\ell,m}^s + \alpha_{\ell',m'}^{s'}\right)\right]}{\omega_L + (\omega_\ell + \omega_{\ell'})} \right) \right\}.
\end{aligned}
$$

For $\omega_L^2 = (\omega_\ell + \omega_{\ell'})^2$:

$$
\begin{aligned}
&{}_2S_L(\ell,m,s;\ell',m',s';t) \\
&= \left\{ - \frac{t}{2(\omega_\ell + \omega_{\ell'})} \cos\left[(\omega_\ell + \omega_{\ell'})t + \left(\alpha_{\ell,m}^s + \alpha_{\ell',m'}^{s'}\right)\right] \right. \qquad (119) \\
&\left. + \frac{1}{4\omega_L(\omega_\ell + \omega_{\ell'})} \left(\sin\left[\omega_L t + \left(\alpha_{\ell,m}^s + \alpha_{\ell',m'}^{s'}\right)\right] + \sin\left[\omega_L t - \left(\alpha_{\ell,m}^s + \alpha_{\ell',m'}^{s'}\right)\right] \right) \right\}.
\end{aligned}
$$

There are two cases for ${}_3S_L$, as follows.

Case 1. For $\omega_L^2 \neq (\omega_\ell - \omega_{\ell'})^2$ and $(\omega_\ell - \omega_{\ell'}) \neq 0$:

$$
\begin{aligned}
&{}_3S(\ell,m,s;\ell',m',s';t) \\
&= \left\{ \frac{\sin\left[(\omega_\ell - \omega_{\ell'})t + \left(\alpha_{\ell,m}^s - \alpha_{\ell',m'}^{s'}\right)\right]}{\omega_L^2 - (\omega_\ell - \omega_{\ell'})^2} \right. \qquad (120a) \\
&\left. - \frac{1}{2\omega_L} \left(\frac{\sin\left[\omega_L t + \left(\alpha_{\ell,m}^s - \alpha_{\ell',m'}^{s'}\right)\right]}{\omega_L - (\omega_\ell - \omega_{\ell'})} - \frac{\sin\left[\omega_L t - \left(\alpha_{\ell,m}^s - \alpha_{\ell',m'}^{s'}\right)\right]}{\omega_L + (\omega_\ell - \omega_{\ell'})} \right) \right\}.
\end{aligned}
$$

Case 2. For $\omega_L^2 \neq (\omega_\ell - \omega_{\ell'})^2$ and $(\omega_\ell - \omega_{\ell'}) = 0$:

$$
{}_3S_L(\ell,m,s;\ell',m',s';t)
$$

$$
-\left\{\frac{1}{\omega_L^2}\sin\left(\alpha^s_{\ell,m} - \alpha^{s'}_{\ell',m'}\right) - \frac{1}{\omega_L^2}\cos\omega_L t\ \sin\left(\alpha^s_{\ell,m} - \alpha^{s'}_{\ell',m'}\right)\right\}. \tag{120b}
$$

There are two cases for ${}_4S_L$, as follows.

Case 1. For $\omega_L^2 = (\omega_\ell - \omega_{\ell'})^2$ and $(\omega_\ell - \omega_{\ell'}) \neq 0$:

$$
{}_4S_L(\ell,m,s;\ell',m',s';t)
$$

$$
=\left\{-\frac{t}{2(\omega_\ell - \omega_{\ell'})}\cos\left[(\omega_\ell - \omega_{\ell'})t + \left(\alpha^s_{\ell,m} - \alpha^{s'}_{\ell',m'}\right)\right]\right. \tag{121a}
$$

$$
\left.+\frac{1}{4\omega_L(\omega_\ell - \omega_{\ell'})}\left(\sin\left[\omega_L t + \left(\alpha^s_{\ell,m} - \alpha^{s'}_{\ell',m'}\right)\right] + \sin\left[\omega_L t - \left(\alpha^s_{\ell,m} - \alpha^{s'}_{\ell',m'}\right)\right]\right)\right\}.
$$

Case 2. For $\omega_L^2 = (\omega_\ell - \omega_{\ell'})^2$ and $(\omega_\ell - \omega_{\ell'}) = 0$:

$$
{}_4S_L(\ell,m,s;\ell',m',s';t)
$$

$$
=\left\{-\frac{t}{2(\omega_\ell - \omega_{\ell'})}\cos\left[(\omega_\ell - \omega_{\ell'})t + \left(\alpha^s_{\ell,m} - \alpha^{s'}_{\ell',m'}\right)\right]\right. \tag{121b}
$$

$$
\left.+\frac{1}{4\omega_L(\omega_\ell - \omega_{\ell'})}\left(\sin\left[\omega_L t + \left(\alpha^s_{\ell,m} - \alpha^{s'}_{\ell',m'}\right)\right] + \sin\left[\omega_L t - \left(\alpha^s_{\ell,m} - \alpha^{s'}_{\ell',m'}\right)\right]\right)\right\}.
$$

The solutions of Eqs. (113) and (115) constructed with the aid of results in Eqs. (116), (117a), and (117b) can now be expressed as

$$
{}_2p^S_{L,M}(t) = \sum_{\Lambda} \sum_{\ell,m,s,\lambda} \sum_{\ell',m',s',\lambda'} E_{L,M,S,\Lambda}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0)
$$

$$
\times\ {}_1p^s_{\ell,m}(0)\ {}_1p^{s'}_{\ell',m'}(0)\ T_L(\ell,m,s;\ell',m',s';t)\,, \tag{122}
$$

Each term in the sum in Eq. (122) consists of three kinds of factors. $E_{L,M,S,\Lambda}$ depends on spatial variables. The quantities ${}_1p^s_{\ell,m}(0)\ {}_1p^{s'}_{\ell',m'}(0)$ depend on which first order waves are excited and on their amplitudes. T_L depends on time and it can be evaluated with the following formulas.

For $\omega_L^2 \neq (\omega_\ell + \omega_{\ell'})^2$ and $\omega_L^2 \neq (\omega_\ell - \omega_{\ell'})^2$:

$$
\begin{aligned}
&T_L(\ell,m,s;\ell',m',s';t) \\
&= {}_1S_L(\ell,m,s;\ell',m',s';t) + {}_3S_L(\ell,m,s;\ell',m',s';t)\,.
\end{aligned} \tag{123a}
$$

For $\omega_L^2 = (\omega_\ell + \omega_{\ell'})^2$ and $\omega_L^2 \neq (\omega_\ell - \omega_{\ell'})^2$:

$$
\begin{aligned}
&T_L(\ell,m,s;\ell',m',s';t) \\
&= {}_2S_L(\ell,m,s;\ell',m',s';t) + {}_3S(\ell,m,s;\ell',m',s';t)\,.
\end{aligned} \tag{123b}
$$

For $\omega_L^2 \neq (\omega_\ell + \omega_{\ell'})^2$ and $\omega_L^2 = (\omega_\ell - \omega_{\ell'})^2$:

$$
\begin{aligned}
&T_L(\ell,m,s;\ell',m',s';t) \\
&= {}_1S_L(\ell,m,s;\ell',m',s';t) + {}_4S_L(\ell,m,s;\ell',m',s';t)\,.
\end{aligned} \tag{123c}
$$

Equations (114), (122) and (123a)-(123c) can be used to completely evaluate the coefficients ${}_2p^S_{L,M}(t)$ in the second order terms for velocity potential in Eq. (109).

Certain features of the time dependent factor T_L in ${}_2p^S_{L,M}(t)$ require further comment. First it should be observed that the exact resonance condition in which a frequency denominator vanishes will hardly ever occur in practice, and may be regarded as accidental because the frequency spectrum for capillary waves in a liquid drop is discrete. However, in those rare instances where a denominator does vanish exactly, the coefficient ${}_2p^S_{L,M}(t)$ contains a term that increases linearly with time. This behavior appears in the functions ${}_2S_L$ in Eq. (119) and ${}_4S_L$ in Eq. (121a). For long enough times, resonances in second order theory would produce waves of such large amplitude that they would dominate the first order waves. The assumptions of perturbation theory on which these results rely would not be met under these conditions. However, the linear growth over some time range is an important feature that may lead to turbulence. This matter is discussed in Sec.VI.

For the discrete spectrum that we are considering, non-resonant conditions apply in almost all cases. Let us focus on ${}_1S_L$ in Eq. (118) as an example of non-resonant behavior. First it should be noted that only terms with sinusoidal time dependence occur in ${}_1S_L$, so that the magnitudes of these terms have finite upper bounds for infinitely long time intervals. However, any spatial state with a given value of L oscillates with at least two different frequencies, viz., ω_L and $(\omega_\ell + \omega_{\ell'})$. Of course, in the formula for ${}_2p^S_{L,M}(t)$) the ℓ and ℓ' are summation indices, and there will be more than two frequencies in any spatial state with index L if more than one first order state is excited initially. The strength of any sinusoidal term is determined by a frequency dependent denominator which is not zero. Although each term is clearly periodic in time, it is noteworthy that for waves that are near resonance, there is some time interval in which the time dependence of ${}_1S_L$ approaches the time dependence of ${}_2S_L$. The length of that time interval of linear growth increases as the near resonant waves approach a resonant condition. This will be demonstrated next.

I. Linear time growth of near-resonant terms

Start with the formula ${}_1S_L$ in Eq. (118) and write the factor involving a denominator as follows:

$$\frac{1}{\omega_L^2-(\omega_\ell+\omega_{\ell'})^2}=\frac{1}{2\omega_L}\left\{\frac{1}{\Delta\omega}+\frac{1}{2(\omega_\ell+\omega_{\ell'})+\Delta\omega}\right\} \tag{124a}$$

$$=\frac{1}{2\omega_L}\left\{\frac{1}{\Delta\omega}+\frac{1}{2(\omega_\ell+\omega_{\ell'})}-\frac{\Delta\omega}{4(\omega_\ell+\omega_{\ell'})^2}+\cdots\right\}, \tag{124b}$$

where $\Delta\omega$ is given by

$$\Delta\omega=\omega_L-(\omega_\ell+\omega_{\ell'}). \tag{124c}$$

Only the two leading terms in a Taylor's series expansion of the second term in Eq. (124a) that are shown explicitly in Eq. (124b) will be retained in what follows.

A trigonometric identity for sin(x-y) allows us to write the first term of Eq. (118) as

$$\sin\left[(\omega_\ell+\omega_{\ell'})t+\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right]$$

$$=\left\{\sin\left[\omega_L t+\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right]\cos[(\Delta\omega)t]-\cos\left[\omega_L t+\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right]\sin[(\Delta\omega)t]\right\} \tag{124d}$$

$$=\left\{\sin\left[\omega_L t+\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right]\left[1-\frac{1}{2}[(\Delta\omega)t]^2+\cdots\right]\right.$$
$$\left.-\cos\left[\omega_L t+\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right]\left[(\Delta\omega)t-\frac{1}{3!}[(\Delta\omega)t]^3+\cdots\right]\right\}, \tag{124e}$$

Here the leading terms in series expansions of sines and cosines have been retained.

Near resonance, where $(\Delta\omega/\omega_L) << 1$, and for times t such that $[(\Delta\omega)t] << 1$, one can retain the leading terms in the expansion of the first term in Eq. (118) and obtain the following approximate equalities:

$$\frac{\sin\left[(\omega_\ell+\omega_{\ell'})t+\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right]}{\omega_L^2-(\omega_\ell+\omega_{\ell'})^2}$$

$$\approx\frac{1}{2\omega_L}\left\{\frac{1}{\Delta\omega}+\frac{1}{2(\omega_\ell+\omega_{\ell'})}\right\}\left\{\sin\left[\omega_L t+\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right]\right. \tag{124f}$$
$$\left.-\cos\left[\omega_L t+\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right][(\Delta\omega)t]\right\}$$

$$\approx\left\{-\frac{1}{2\omega_L}t\cos\left[\omega_L t+\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right]\right.$$
$$\left.+\frac{1}{4\omega_L(\omega_\ell+\omega_{\ell'})}\sin\left[\omega_L t+\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right]+\frac{1}{2\omega_L(\Delta\omega)}\sin\left[\omega_L t+\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right]\right\}. \tag{124g}$$

Similar treatment of the last two terms of Eq. (118) yields the following approximate equality:

$$
\begin{aligned}
&- \frac{1}{2\omega_L}\left(\frac{\sin\left[\omega_L t + \left(\alpha^s_{\ell,m} + \alpha^{s'}_{\ell',m'}\right)\right]}{\omega_L - (\omega_\ell + \omega_{\ell'})} - \frac{\sin\left[\omega_L t - \left(\alpha^s_{\ell,m} + \alpha^{s'}_{\ell',m'}\right)\right]}{\omega_L + (\omega_\ell + \omega_{\ell'})}\right) \\
&\approx - \frac{1}{2\omega_L}\left(\frac{\sin\left[\omega_L t + \left(\alpha^s_{\ell,m} + \alpha^{s'}_{\ell',m'}\right)\right]}{\Delta\omega} - \frac{\sin\left[\omega_L t - \left(\alpha^s_{\ell,m} + \alpha^{s'}_{\ell',m'}\right)\right]}{2(\omega_\ell + \omega_{\ell'})}\right).
\end{aligned}
\tag{124h}
$$

Substituting Eqs. (124g) and (124h) into the formula for ${}_1S_L$ in Eq. (118), one finds that the terms with the small quantity $\Delta\omega$ in their denominators cancel, and one obtains the following result. For $|\Delta\omega / \omega_L| << 1$ and $|\Delta\omega)t| << 1$:

$$
\begin{aligned}
&{}_1S_L(\ell,m,s;\ell',m',s';t) \\
&\approx \left\{- \frac{t}{2\omega_L}\cos\left[\omega_L t + \left(\alpha^s_{\ell,m} - \alpha^{s'}_{\ell',m'}\right)\right]\right. \\
&\left. + \frac{1}{4\omega_L(\omega_\ell + \omega_{\ell'})}\left(\sin\left[\omega_L t + \left(\alpha^s_{\ell,m} + \alpha^{s'}_{\ell',m'}\right)\right] + \sin\left[\omega_L t - \left(\alpha^s_{\ell,m} + \alpha^{s'}_{\ell',m'}\right)\right]\right)\right\}.
\end{aligned}
\tag{124i}
$$

The limiting form of ${}_1S_L$ in Eq. (124i) agrees with the result for ${}_2S_L$ at the exact resonance condition given in Eq. (119) when one takes into account that $\omega_L = \omega_\ell + \omega_{\ell'}$ at resonance in Eq. (119). This result is important for the proposed explanation of intermittency in ripple turbulence discussed in Sec.VI.

J. General results for ${}_2\eta(\theta, \varphi; t)$

Now we shall derive formulas for ${}_2\eta(\theta, \varphi; t)$, the second order contribution to displacement of the free surface of the liquid. In what follows we shall use a prime and a double prime to indicate first and second partial derivatives, respectively, with respect to r. The derivation begins with Eq. (46b), which takes the following form in the new notation. For $r=r_0$:

$$ {}_2\dot{\eta}-{}_2\Phi'-{}_1\Phi''{}_1\eta+\left(\vec{\nabla}_1\eta\right)\cdot\left(\vec{\nabla}_1\Phi\right)=0 \tag{125} $$

Using Eq. (110), one finds

$$ {}_2\dot{\eta}(\theta,\varphi;t)=\sum_{L_1,M_1,S_1} {}_2\dot{z}^{S_1}_{L_1,M_1}(t)\,C^{S_1}_{L_1,M}(\theta,\varphi)\ . \tag{126a} $$

With the aid of Eq. (109), one finds that

$$ {}_2\Phi'(r_0,\theta,\varphi;t)=\sum_{L_1,M_1,S_1} {}_2p^{S_1}_{L_1,M_1}(t)\,R'_{L_1}(r_0)\,C^{S_1}_{L_1,M_1}(\theta,\varphi)\,. \tag{126b} $$

Using a procedure similar to that applied in evaluating P_1 in Eqs. (81b) and (88) and ${}_1D_{\ell,m}$ in Eq. (89), one finds at $r=r_0$:

$$ {}_1\Phi''{}_1\eta=\sum_{L_1,M_1}\ \sum_{\ell,m,s,\lambda}\ \sum_{\ell',m',s',\lambda'}\left(\alpha^{s,\lambda}_{m}\right)^*\alpha^{s',\lambda'}_{m'} \tag{126c} $$
$$ \times\,{}_8D_{L_1,M_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0)\,{}_1z^{s}_{\ell,m}(t)\,{}_1p^{s'}_{\ell',m'}(t)\,Y_{L_1,M_1}(\theta,\varphi)\,, $$

where

$$
{}_8D_{L_1,M_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0)
$$

$$
=\left\{\frac{\alpha}{\rho r_0{}^2}\left[2-L_1(L_1+1)\right]\right\}^{-1}{}_1D_{L_1,M_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0). \tag{126d}
$$

The term $(\nabla_1\eta)\bullet(\nabla\Phi)$ can be evaluated using the same basic procedure as that applied when treating P_7 in Eqs. (81h) and (107) and ${}_7D_{\ell,m}$ in Eq. (108). The result is

$$
-(\vec{\nabla}_1\eta)\cdot(\vec{\nabla}_1\Phi)=\sum_{L_1,M_1}\sum_{\ell,m,s,\lambda}\sum_{\ell',m',s',\lambda'}\left(\alpha_m^{s,\lambda}\right)^*\alpha_{m'}^{s',\lambda'}
$$

$$
\times\,{}_9D_{L_1,M_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0)\,{}_1z_{\ell,m}^s(t)\,{}_1p_{\ell',m'}^{s'}(t)Y_{L_1,M_1}(\theta,\varphi), \tag{126e}
$$

where

$$
{}_9D_{L_1,M_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0)
$$

$$
=\left\{\frac{\alpha}{\rho r_0{}^2}\left[2-L_1(L_1+1)\right]\right\}^{-1}{}_7D_{L_1,M_1}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0). \tag{126f}
$$

Next substitute Eqs. (126a)-(126c) and (126e) into (125). Eliminate ${}_1z_{\ell,m}^s(t)$ from the subsequent equation with the aid of Eq. (77a). Then multiply that result by $C_{L,M}^S(\theta,\varphi)$) and integrate the subsequent expression over a unit sphere. Then use Eqs. (60), (61), and (63) and solve for ${}_2\dot{z}_{\ell,m}^s(t)$. The result is

$$
\begin{aligned}
{}_2\dot{z}^S_{L,M}(t) = \Big\{ & {}_2p^S_{L,M}(t)\ R'_L(r_0) \\
&+\sum_{\Lambda}\ \sum_{\ell,m,s,\lambda}\ \sum_{\ell',m',s',\lambda'} F_{L,M,S,\Lambda}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0) \\
&\times {}_1p^s_{\ell,m}(0)\ {}_1p^{s'}_{\ell',m'}(0)\ \sin\left(\omega_\ell t+\alpha^s_{\ell,m}\right)\cos\left(\omega_{\ell'}t+\alpha^{s'}_{\ell',m'}\right)\Big\},
\end{aligned}
\tag{127}
$$

where

$$
\begin{aligned}
&F_{L,M,S,\Lambda}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0) \\
&=\left[\frac{R'_\ell(r_0)}{N(L,M,S)\omega_\ell}\right]\left(a^{S,\Lambda}_M\right)^*\left(a^{s,\lambda}_m\right)^* a^{s',\lambda'}_{m'} \sum_{8\le j\le 9} {}_jD_{L,(\Lambda M)}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0)\,.
\end{aligned}
\tag{128}
$$

Next combine Eqs. (122) and (127) and obtain

$$
\begin{aligned}
&{}_2\dot{z}^S_{L,M}(t) \\
&=\sum_{\Lambda}\ \sum_{\ell,m,s,\lambda}\ \sum_{\ell',m',s',\lambda'} {}_1p^s_{\ell,m}(0)\ {}_1p^{s'}_{\ell',m'}(0) \\
&\times\Big\{R'_\ell(r_0)\ E_{L,M,S,\Lambda}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0)\ T_L(\ell,m,s;\ell',m',s';t) \\
&+\ F_{L,M,S,\Lambda}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0)\sin\left(\omega_\ell t+\alpha^s_{\ell,m}\right)\cos\left(\omega_{\ell'}t+\alpha^{s'}_{\ell',m'}\right)\Big\}.
\end{aligned}
\tag{129}
$$

A formula for ${}_2z^s_{\ell,m}(t)$ can be obtained by integrating Eq. (129) with respect to time. The indefinite integral is sufficient to find the time dependence since the initial values ${}_2z^s_{\ell,m}(0)$have not been specified. The result is

$$
\begin{aligned}
&{}_2z^{S}_{L,M}(t) \\
&= \sum_{\Lambda} \sum_{\ell,m,s,\lambda} \sum_{\ell',m',s',\lambda'} {}_1p^{s}_{\ell,m}(0)\ {}_1p^{s'}_{\ell',m'}(0) \\
&\times \Big\{ R'_{\ell}(r_0)\ E_{L,M,S,\Lambda}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0)\ U_L(\ell,m,s;\ell',m',s';t) \\
&+ F_{L,M,S,\Lambda}(\ell,m,s,\lambda;\ell',m',s',\lambda';r_0)\, W(\ell,m,s;\ell',m',s';t) \Big\}.
\end{aligned} \tag{130}
$$

In an abbreviated notation where some of the arguments of functions are suppressed, we have

$$U_L = \int^{t} dt' \, T_L(t') \tag{131}$$

$$W = \int^{t} dt' \, \sin\!\left(\omega_{\ell} t' + \alpha^{s}_{\ell,m}\right) \cos\!\left(\omega_{\ell'} t' + \alpha^{s'}_{\ell',m'}\right). \tag{132}$$

It is useful to introduce functions ${}_jV_L$ that correspond to ${}_jS_L$ in Eqs. (118)-(121) in order to efficiently represent U_L. For each value of j , where $1 \le j \le 9$, we have

$$
\begin{aligned}
&{}_jV_L(\ell,m,s;\ell',m',s';t) \\
&= \int^{t} dt' \ {}_jS_L(\ell,m,s;\ell',m',s';t).
\end{aligned} \tag{133}
$$

For $\omega_L^2 \neq (\omega_{\ell} + \omega_{\ell'})^2$:

$$
{}_1V_L(\ell,m,s;\ell',m',s';t)
$$

$$
= -\left\{\frac{\cos\left[(\omega_\ell+\omega_{\ell'})t+\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right]}{(\omega_\ell+\omega_{\ell'})\left[\omega_L^2-(\omega_\ell+\omega_{\ell'})^2\right]}\right. \tag{134}
$$

$$
\left.-\frac{1}{2\omega_L}\left(\frac{\cos\left[\omega_L t+\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right]}{\omega_L\left[\omega_L-(\omega_\ell+\omega_{\ell'})\right]}-\frac{\cos\left[\omega_L t-\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right]}{\omega_L\left[\omega_L+(\omega_\ell+\omega_{\ell'})\right]}\right)\right\}.
$$

For $\omega_L^2=(\omega_\ell+\omega_{\ell'})^2$:

$$
{}_2V_L(\ell,m,s;\ell',m',s';t)
$$

$$
= -\left\{\frac{1}{2(\omega_\ell+\omega_{\ell'})^3}\left[\left((\omega_\ell+\omega_{\ell'})t+\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right)\sin\left[(\omega_\ell+\omega_{\ell'})t+\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right]\right.\right.
$$

$$
\left.+\cos\left[(\omega_\ell+\omega_{\ell'})t+\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right]-\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\sin\left[(\omega_\ell+\omega_{\ell'})t+\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right]\right] \tag{135}
$$

$$
\left.+\frac{1}{4\omega_L^2(\omega_\ell+\omega_{\ell'})}\left[\left[\omega_L t+\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right]+\cos\left[\omega_L t-\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right]\right]\right\}
$$

There are two cases for ${}_3V_L$, as follows:

Case 1. For $\omega_L^2\neq(\omega_\ell-\omega_{\ell'})^2$ and $(\omega_\ell-\omega_{\ell'})\neq 0$:

$$
{}_3V_L(\ell,m,s;\ell',m',s';t)
$$

$$
= -\left\{\frac{\cos\left[(\omega_\ell-\omega_{\ell'})t+\left(\alpha^s_{\ell,m}-\alpha^{s'}_{\ell',m'}\right)\right]}{(\omega_\ell-\omega_{\ell'})\left[\omega_L^2-(\omega_\ell-\omega_{\ell'})^2\right]}\right. \tag{136a}
$$

$$
\left.-\frac{1}{2\omega_L{}^2}\left(\frac{\cos\left[\omega_L t+\left(\alpha^s_{\ell,m}-\alpha^{s'}_{\ell',m'}\right)\right]}{\omega_L-(\omega_\ell-\omega_{\ell'})}-\frac{\cos\left[\omega_L t-\left(\alpha^s_{\ell,m}-\alpha^{s'}_{\ell',m'}\right)\right]}{\omega_L+(\omega_\ell-\omega_{\ell'})}\right)\right\}.
$$

Case 2. For $\omega_L^2 \neq (\omega_\ell - \omega_{\ell'})^2$ and $(\omega_\ell - \omega_{\ell'}) = 0$:

$$
{}_3V_L(\ell,m,s;\ell',m',s';t)
= \left\{ \frac{1}{\omega_L^2} \sin\left(\alpha^s_{\ell,m} - \alpha^{s'}_{\ell',m'}\right) - \frac{1}{\omega_L^2} \cos\omega_L t \sin\left(\alpha^s_{\ell,m} - \alpha^{s'}_{\ell',m'}\right) \right\}. \tag{136b}
$$

There are two cases for ${}_4V_L$, as follows:

Case 1. For $\omega_L^2 = (\omega_\ell - \omega_{\ell'})^2$ and $(\omega_\ell - \omega_{\ell'}) \neq 0$:

$$
\begin{aligned}
{}_4V_L(\ell,m,s;\ell',m',s';t)
&= - \Bigg\{ \frac{1}{2(\omega_\ell - \omega_{\ell'})^3} \Bigg[\left((\omega_\ell - \omega_{\ell'})t + \left(\alpha^s_{\ell,m} - \alpha^{s'}_{\ell',m'}\right)\right) \sin\left[(\omega_\ell - \omega_{\ell'})t + \left(\alpha^s_{\ell,m} - \alpha^{s'}_{\ell',m'}\right)\right] \\
&+ \cos\left[(\omega_\ell - \omega_{\ell'})t + \left(\alpha^s_{\ell,m} - \alpha^{s'}_{\ell',m'}\right)\right] - \left(\alpha^s_{\ell,m} - \alpha^{s'}_{\ell',m'}\right) \sin\left[(\omega_\ell - \omega_{\ell'})t + \left(\alpha^s_{\ell,m} - \alpha^{s'}_{\ell',m'}\right)\right] \Bigg] \\
&+ \frac{1}{4\omega_L^2(\omega_\ell - \omega_{\ell'})} \left[\cos\left[\omega_L t + \left(\alpha^s_{\ell,m} - \alpha^{s'}_{\ell',m'}\right)\right] + \cos\left[\omega_L t - \left(\alpha^s_{\ell,m} - \alpha^{s'}_{\ell',m'}\right)\right] \right] \Bigg\}.
\end{aligned} \tag{137a}
$$

Case 2. For $\omega_L^2 = (\omega_\ell - \omega_{\ell'})^2$ and $(\omega_\ell - \omega_{\ell'}) = 0$:

$$
{}_4V_L(\ell,m,s;\ell',m',s';t) = 0\,. \tag{137b}
$$

U_L can now be specified in terms of non-overlapping regions of frequency, just as was done previously for T_L. The formulas are given next.

For $\omega_L^2 \neq (\omega_\ell + \omega_{\ell'})^2$ and $\omega_L^2 \neq (\omega_\ell - \omega_{\ell'})^2$:

$$U_L(\ell,m,s;\ell',m',s';t)={}_1V_L(\ell,m,s;\ell',m',s';t)+{}_3V_L(\ell,m,s;\ell',m',s';t)\,. \tag{138a}$$

For $\omega_L^2=(\omega_\ell+\omega_{\ell'})^2$ and $\omega_L^2\neq(\omega_\ell-\omega_{\ell'})^2$:

$$U_L(\ell,m,s;\ell',m',s';t)={}_2V_L(\ell,m,s;\ell',m',s';t)+{}_3V_L(\ell,m,s;\ell',m',s';t)\,. \tag{138b}$$

For $\omega_L^2\neq(\omega_\ell+\omega_{\ell'})^2$ and $\omega_L^2=(\omega_\ell-\omega_{\ell'})^2$:

$$U_L(\ell,m,s;\ell',m',s';t)={}_1V_L(\ell,m,s;\ell',m',s';t)+{}_4V_L(\ell,m,s;\ell',m',s';t)\,. \tag{138c}$$

This completes the evaluation of U_L in Eq. (130). The next task is to evaluate the function W in Eqs.(130) and (132). Using a trigonometric identity, one can write the integrand of Eq. (132) as follows.

For $(\omega_\ell-\omega_{\ell'})\neq 0$:

$$\begin{aligned}&\sin\left(\omega_\ell t+\alpha^s_{\ell,m}\right)\cos\left(\omega_{\ell'}t+\alpha^{s'}_{\ell',m'}\right)\\&=\frac{1}{2}\left\{\sin\left[(\omega_\ell+\omega_{\ell'})t+\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right]+\sin\left[(\omega_\ell-\omega_{\ell'})t+\left(\alpha^s_{\ell,m}-\alpha^{s'}_{\ell',m'}\right)\right]\right\}.\end{aligned} \tag{139a}$$

For $(\omega_\ell-\omega_{\ell'})=0$:

$$\begin{aligned}&\sin\left(\omega_\ell t+\alpha^s_{\ell,m}\right)\cos\left(\omega_{\ell'}t+\alpha^{s'}_{\ell',m'}\right)\\&=\frac{1}{2}\left\{\sin\left[(\omega_\ell+\omega_{\ell'})t+\left(\alpha^s_{\ell,m}+\alpha^{s'}_{\ell',m'}\right)\right]+\sin\left(\alpha^s_{\ell,m}-\alpha^{s'}_{\ell',m'}\right)\right\}.\end{aligned} \tag{139b}$$

Substitute Eq. (139a) into (132) and then integrate. After that substitute Eq. (139b) into (132) and integrate. One finds the following results

For $(\omega_\ell - \omega_{\ell'}) \neq 0$:

$$W(\ell,m,s;\ell',m',s';t)$$
$$= -\frac{1}{2}\left\{\frac{\cos\left[(\omega_\ell + \omega_{\ell'})t + \left(\alpha^{s}_{\ell,m} + \alpha^{s'}_{\ell',m'}\right)\right]}{\omega_\ell + \omega_{\ell'}} + \frac{\cos\left[(\omega_\ell - \omega_{\ell'})t + \left(\alpha^{s}_{\ell,m} - \alpha^{s'}_{\ell',m'}\right)\right]}{\omega_\ell - \omega_{\ell'}}\right\}. \tag{140a}$$

For $(\omega_\ell - \omega_{\ell'}) = 0$:

$$W(\ell,m,s;\ell',m',s';t)$$
$$= -\frac{1}{2}\left\{\frac{\cos\left[(\omega_\ell + \omega_{\ell'})t + \left(\alpha^{s}_{\ell,m} + \alpha^{s'}_{\ell',m'}\right)\right]}{\omega_\ell + \omega_{\ell'}} + \frac{1}{2}t\sin\left(\alpha^{s}_{\ell,m} - \alpha^{s'}_{\ell',m'}\right)\right\}. \tag{140b}$$

This completes the derivation of formulas needed to evaluate ${}_2z^{s}_{\ell,m}(t)$ in Eq. (130). Then ${}_2\eta(\theta,\varphi;t)$ can be evaluated using Eq. (110).

VI. DISCUSSION OF RESULTS

A. Canonical formalism

The variational theory of a hydrodynamic system with moving boundaries involves a number of subtle problems. Therefore it is important to clearly describe the assumptions and mathematical procedure and demonstrate the self-consistency of the method by presenting details, as in Sec.II. In earlier work, Zakharov[15] treated the exact theory of finite amplitude waves on a fluid surface in planar geometry. He showed that the equations for the time dependence of the surface could be expressed in canonical form. However the details of Zakharov's method differ in non-trivial ways from the corresponding elements of the present theory.

The essence of the difference between the two methods is the condition under which the variation $\delta\Phi$ is taken. In the present theory, the variation $\delta\Phi$ of the velocity potential in the interior of the liquid as well as on its boundary is taken at any instant of time with the surface held fixed at its actual path position for that time instant. An important consequence of this prescription for varying the velocity potential is that the canonical momentum and canonical coordinate are manifestly treated as independent variables in terms of variables evaluated at the drop surface.

In Zakharov's treatment, Φ evaluated at the fluid surface, which Zakharov then calls Ψ, is a generalized coordinate and the surface displacement η is a generalized momentum. This particular difference between Zakharov's treatment and the present treatment is not of fundamental importance since it is known from general theory that a canonical transformation can interchange canonical momenta and coordinates. However, there is a fundamental difference in the two theories that results from Zakharov considering the variation $\delta\Phi$ to be a function of $\delta\eta$. Since $\delta\Phi$ evaluated at the drop's surface is a generalized coordinate in Zakharov's theory, one encounters the seemingly paradoxical situation in which the canonical coordinate Ψ is a function of the canonical momentum η, and they are therefore not independent variables. This appears to conflict with general principles of canonical formalism. This matter needs further clarification in Zakharov's method.

Theoretical treatments of surface waves and wave turbulence have utilized the properties of canonical variables in a large body of published work. This provided a strong incentive to establish that the present theory could be expressed in canonical form. Furthermore, with the aid of canonical equations it was relatively simple to demonstrate that the energy of the system is a constant of the motion in the exact theory. This was especially important in establishing self-consistency of this method because in low order perturbation theory of the almost spherical drop, energy is not a constant of the motion. This result was developed in detail in Sec.V. This puzzling result should be a proper subject for further work and comment. As stated in Sec.V, this difficulty does not occur in planar geometry where almost flat surfaces are treated.

Additionally, it should be noted that the equations of motion thru first order for the surface of an almost spherical drop obtained from perturbation expansions of the exact equation of motion cannot be obtained as canonical equations based on the perturbation expansion of the exact Hamiltonian thru second order. This condition is related to the geometric factor r^2 in the differential of the surface element df, where $df = \gamma r^2(\theta, \varphi; t) \sin\theta d\theta d\varphi$. It is noteworthy that if the Hamiltonian density in Eq. (52) were multiplied by $(1-\eta/r_0) \approx r_0/r(\theta,\varphi;t)$, then energy evaluated thru quadratic terms in perturbation theory would be constant in time. However, a justification for this step has not yet been found.

Absence of the canonical property is related to the difficulty with the energy not being a constant of the motion at this level of perturbation theory. But just as important, absence of the canonical property undermines application of the canonical transformation method in treating surface turbulence in liquid drops in perturbation theory. Canonical formalism has frequently been used in applying perturbation methods in theories of turbulence in surface waves on almost flat surfaces.

B. L=1 tesseral harmonic terms

In Appendix C it is shown that the presence of an L=1 tesseral harmonic contribution to surface displacement would imply displacement of the center of mass of an almost spherical drop. In the model considered in this paper, it was specified that the L=1 terms must be absent in all orders of perturbation theory. However, even if the center of mass were not constrained to be stationary, and even if L=1 terms were absent in first order perturbation theory, there is a possibility that L=1 terms would occur in second order because of nonlinear interactions involving frequencies $\omega_L = \omega_\ell - \omega_{\ell'} = 0$, where $\ell = \ell' \neq 1$. Analysis shows that if such L=1 terms did occur, they would have a t^2 time dependence in the velocity potential. A proper method for dealing with that situation would require further analysis.

In the early stages of development of the present theory, it was planned that the model would consist of a liquid mantle surrounding a thin solid, rigid shell. For example, the shell could be imagined as a ping pong ball, say having outer radius r_c. Then a liquid drop would occur as a limiting case in which r_c tends to zero. If one were to disregard L=1 terms, the mathematical theory of this liquid mantle would differ from the present model just in the formula for the radial function $R_\ell(r)$ in the expansion of the velocity potential. The new radial function would satisfy a boundary condition that the radial component of the velocity vanish at r_c instead of at r=0. The liquid mantle model was abandoned because it appeared that L=1 terms that involve motion of the mantle with respect to the rigid shell might occur even when the center of mass remained stationary. The t^2 time dependence in velocity potential for L=1 terms that might then occur in second order perturbation theory would require analysis and interpretation beyond that which has been considered so far.

C. Intermittency in wave turbulence

A full description of the path to turbulence would require calculations that usually involve statistical methods or dimensional analysis.[8,9,21] Nevertheless, the second order perturbation results in this paper that can describe an initial step on a path to turbulence suggests a physical mechanism that may be responsible for intermittency, that is, occurrence of large amplitude fluctuations in the surface over relatively small areas.[3,11]

The proposed explanation for intermittency can be understood by considering a simple situation in which two first order capillary waves are excited at high amplitude, and where the sum of the first order frequencies, $(\omega_\ell + \omega_{\ell'})$, is near a resonant frequency ω_L in the first order spectrum. According to Eqs. (122) and (123a), the time dependent factor

T_L in the second order amplitude ${}_2p^S_{L,M}(t)$ will include a term ${}_lS_L$. Equation (118) shows that ${}_lS_L$ includes terms that oscillate with frequencies $(\omega_\ell + \omega_{\ell'})$ and ω_L, whose difference is $\Delta\omega = \omega_L - (\omega_\ell + \omega_{\ell'})$, as in Eq. (124c). In Eqs. (124a)-(124i) it was shown that the two terms with small denominators in ${}_lS_L$ combine to produce an envelope that grows linearly in time, and it was noted there that this growth persists for a time t whose length is determined by the condition $|(\Delta\omega)t| << 1$. In regions near the maximum in amplitude of the spatial factor for the state with frequency ω_L, the second order amplitude may increase to large values during that time period. Eventually, for longer times t, the inequality above will not be satisfied, and linear time growth will cease. The growth may be said to roll over. According to Eq. (118), for longer times there will still be two oscillation frequencies in the spatial state for ω_L. Proximity to the resonance condition will determine the denominators in Eq. (118) that dictate how large the combined amplitude in that spatial state grows. For some even longer time, the two oscillations will be out of phase and the combined amplitude will be small. However, for still later times the two oscillatory terms will achieve almost the same phase relation they had at t=0. Then the linear time growth that occurred near t=0 will occur again, and the combined amplitude near the maximum for the spatial state corresponding to ω_L will grow to a large value.

To see this in more detail, consider the ratio $R = (\omega_\ell + \omega_{\ell'})/\omega_L$. Let T_1 and T_2 be the periods corresponding to $(\omega_\ell + \omega_{\ell'})$ and ω_L, respectively. Then $R=T_2/T_1$. Suppose R=1.001, which is a near-resonance condition. In a time interval $\Delta t = (1001)T_1 = (1000)T_2$ both oscillatory terms will have completed an integral number of cycles, and the sinusoidal factors in the near-resonant terms in Eq. (118) will have returned to the values they had near t=0. Then the combined effect of these two oscillations will be linear growth in time, just as it was near t=0. Clearly these conditions will also occur at later times, giving the appearance of intermittent large fluctuations in amplitude near the maximum of the spatial factor for ω_L.

D. Low Energy Nuclear Physics

Rayleigh waves in the liquid drop model of an atomic nucleus have provided a basis for predicting fairly accurately the threshold of stability with respect to fission.[22] The calculations for stability in that model take into account the weakening of the effect of the ordinary surface tension in the Weizacker semi-empirical mass formula by the Coulomb effect associated with electrical charge in the nucleus. The threshold of stability is associated with the condition that the surface tension factor in the frequency for Rayleigh waves in the L=2 mode becomes negative. Only a small perturbation from the outside is necessary to induce fission in a nucleus that is just below the threshold when in its ground state. The synthesis of the liquid drop model with the nuclear shell model based on independent particle motion provides a formalism [23,24] for calculating γ-ray emission from nuclei in which there are collective motions such as Rayleigh waves.

These observations lead us to speculate that turbulence, like that in a stormy sea, may be generated on the surface of an atomic nucleus that is somewhat below the fission threshold where the effective surface tension is small and the nucleus can be easily distorted by external perturbations. Finite amplitude capillary waves that are excited directly may produce other waves that grow in time due to nonlinear interactions, just as in a water drop. The initial excitation may be due to a near miss by a charged particle, for example. The footprints of the second order waves may be sought in the harmonic structure of γ-ray spectra. For long periods of excitation the footprints of turbulence and intermittency in the wave motion may be observable. This creates an interesting situation in which turbulence may occur in a nuclear "drop" that is strongly influenced by quantum behavior.

Experiments on levitated electrically charged liquid drops by Rhim et al [16] have clearly exhibited amplitude growth of shape oscillations induced by perturbation of the drop. Furthermore, the measured power spectrum of those oscillations shows harmonic structure consistent with nonlinear interactions of finite amplitude waves. Although the present theory only accounts directly for the second harmonic, it is expected that a cascade to higher harmonics will be generated as the power in the low harmonics increases. These experimental results for drops support the speculations on behavior of a perturbed atomic nucleus described above.

ACKNOWLEDGMENTS

I wish to acknowledge the special contributions of Dr. Martin Barmatz to this research and thank him for many clarifying discussions and critical comments. Valuable discussions with Prof. Seth Putterman and Dr. William Wright are also acknowledged. The research described in this paper was carried out at the Jet Propulsion Laboratory, California Institute of Technology, under contract with the National Aeronautics and Space Administration.

APPENDIX A

Potential energy contributions to the Hamiltonian and to Cauchy's Integral in spherical geometry

The Hamiltonian contains two potential energy terms, H_b associated with surface tension, and H_c associated with externally applied pressure. Formulas for these terms and their variational derivatives expressed in spherical coordinates and variables are developed in this appendix. An assumption of local mechanical equilibrium is used to identify terms in Cauchy's integral of the Euler equation of motion.

A. Surface tension term H_b in the potential energy

The energy H_b associated with surface tension α is proportional to the area of the free surface S, and is given by the formula

$$H_b = \alpha \iint_s df \,, \tag{A1}$$

where df is an element of surface area. An explicit expression for H_b will be derived next.

The shape of the free surface at time t is specified by $\vec{r}(\theta, \varphi; t)$, where

$$\vec{r}(\theta, \varphi; t) = \hat{r}\, r(\theta, \varphi; t)\,; \tag{A2a}$$

$$r(\theta, \varphi; t) = r_0 + \eta(\theta, \varphi; t)\,. \tag{A2b}$$

Any differential line element tangent to the surface can be expressed as

$$d\vec{r} = (dr)\hat{r} + (r d\theta)\hat{\theta} + (r \sin\theta d\varphi)\hat{\varphi}\,. \tag{A3a}$$

$$= \left(\frac{\partial \eta}{\partial \theta} d\theta + \frac{\partial \eta}{\partial \varphi} d\varphi \right) \hat{r} + (r d\theta)\hat{\theta} + (r \sin\theta d\varphi)\hat{\varphi}\,. \tag{A3b}$$

Two non-collinear differential line elements $\vec{\rho}_1$ and $\vec{\rho}_2$, tangent to the surface, can be found from Eq. (A3b) by holding φ constant and then holding θ constant, respectively. This yields

$$\vec{\rho}_1 = \left(\frac{\partial \eta}{\partial \theta} \hat{r} + r\hat{\theta} \right) d\theta \tag{A4a}$$

$$\vec{\rho}_2 = \left(\frac{\partial \eta}{\partial \varphi} \hat{r} + r \sin\theta \hat{\varphi} \right) d\varphi\,. \tag{A4b}$$

A differential surface element $d\vec{f}$ normal to the surface and defined by $\vec{\rho}_1$ and $\vec{\rho}_2$ is given by

$$d\vec{f} = \vec{\rho}_1 \times \vec{\rho}_2 \tag{A5a}$$

$$= \left(\hat{r} - \vec{\nabla}\eta\right) r^2(\theta,\varphi;t) \sin\theta \, d\theta \, d\varphi \tag{A5b}$$

$$= \hat{n} \, df , \tag{A5c}$$

where

$$\vec{\nabla}\eta(\theta,\varphi;t) = \frac{1}{r(\theta,\varphi;t)} \left(\hat{\theta} \frac{\partial\eta(\theta,\varphi;t)}{\partial\theta} + \hat{\varphi} \frac{1}{\sin\theta} \frac{\partial\eta(\theta,\varphi;t)}{\partial\varphi} \right) \tag{A6a}$$

$$df = \left[1 + \left(\vec{\nabla}\eta\right)^2 \right]^{1/2} r^2(\theta,\varphi;t) \sin\theta \, d\theta \, d\varphi \tag{A6b}$$

$$\hat{n} = \left(\hat{r} - \vec{\nabla}\eta\right) / \gamma \tag{A6c}$$

$$\gamma = \left[1 + \left(\vec{\nabla}\eta\right)^2 \right]^{1/2} . \tag{A6d}$$

Combining the results in Eqs. (A1)-(A6d), one finds

$$H_b = \alpha \int_0^{2\pi} \int_0^{\pi} \left\{ \left[1 + \left(\vec{\nabla}\eta\right)^2 r^2 \right] \right\}_{r=r_0+\eta(\theta,\varphi;t)} \sin\theta \; d\theta \, d\varphi \tag{A7a}$$

$$= \alpha \int_0^{2\pi} \int_0^{\pi} \left\{ \left[r^2 + \left(\frac{\partial\eta}{\partial\theta}\right)^2 + \frac{1}{\sin^2\theta} \left(\frac{\partial\eta}{\partial\varphi}\right)^2 \right]^{1/2} r \right\}_{r=r_0+\eta(\theta,\varphi;t)} \sin\theta \, d\theta \, d\varphi . \tag{A7b}$$

Next calculate the variation of H_b thru first order when $\eta \rightarrow \eta + \delta\eta$. Using Eq. (A7b) one finds

$$\delta H_b^\eta = H_b[\eta+\delta\eta] - H_b[\eta] \tag{A8a}$$

$$= \alpha \int_0^{2\pi} \int_0^{\pi} \left\{ r\left(\frac{1}{\gamma}+\gamma\right)\delta\eta + \frac{1}{\gamma}\left(\frac{\partial\eta}{\partial\theta}\right)\left(\frac{\partial\delta\eta}{\partial\theta}\right) + \frac{1}{\gamma\sin^2\theta}\left(\frac{\partial\eta}{\partial\varphi}\right)\left(\frac{\partial\delta\eta}{\partial\varphi}\right)\right\}_{r=r_0+\eta(\theta,\varphi;t)} \sin\theta\, d\theta\, d\varphi. \tag{A8b}$$

Integrate the second and third terms in Eq. (A8b) by parts using

$$\frac{\partial}{\partial\theta}\left[\left(\frac{\sin\theta}{\gamma}\frac{\partial\eta}{\partial\theta}\right)\delta\eta\right] = \frac{\sin\theta}{\gamma}\frac{\partial\eta}{\partial\theta}\frac{\partial\delta\eta}{\partial\theta} + \left[\frac{\partial}{\partial\theta}\left(\frac{\sin\theta}{\gamma}\frac{\partial\eta}{\partial\theta}\right)\right]\delta\eta \tag{A9a}$$

$$\frac{\partial}{\partial\varphi}\left[\left(\frac{1}{\gamma}\frac{\partial\eta}{\partial\varphi}\right)\delta\eta\right] = \left[\frac{\partial}{\partial\varphi}\left(\frac{1}{\gamma}\frac{\partial\eta}{\partial\varphi}\right)\right]\delta\eta + \left(\frac{1}{\gamma}\frac{\partial\eta}{\partial\varphi}\right)\frac{\partial\,\delta\eta}{\partial\varphi}. \tag{A9b}$$

The end point contributions to the integral vanish, and one finds

$$\delta H_b^\eta = \alpha \int_0^{2\pi} \int_0^{\pi} \left\{ \left(\frac{1}{r}\left(\frac{1}{\gamma}+\gamma\right) - \frac{1}{r^2\sin\theta'}\left[\frac{\partial}{\partial\theta'}\left(\frac{\sin\theta'}{\gamma}\frac{\partial\eta}{\partial\theta'}\right) + \frac{\partial}{\partial\varphi'}\left(\frac{1}{\gamma}\frac{\partial\eta}{\partial\varphi'}\right)\right]\right) \times(\delta\eta)r^2\right\}_{r=r_0+\eta(\theta',\varphi';t)} \sin\theta'\, d\theta'\, d\varphi'. \tag{A10}$$

Use the basic variational derivative

$$\frac{\partial\eta(\theta',\varphi';t)}{\partial\eta(\theta,\varphi;t)} = \frac{1}{r^2(\theta,\varphi;t)\sin\theta}\delta(\theta-\theta')\delta(\varphi-\varphi') \tag{A11}$$

to calculate $\delta H_b / \delta\eta$. After integrating over Dirac δ functions, one finds

$$\frac{\delta H_b}{\delta\eta(\theta,\varphi;t)} = \alpha\left\{\frac{1}{r}\left(\frac{1}{\gamma}+\gamma\right) - \frac{1}{r^2\sin\theta}\left[\frac{\partial}{\partial\theta}\left(\frac{\sin\theta}{\gamma}\frac{\partial\eta}{\partial\theta}\right) + \frac{\partial}{\partial\varphi}\left(\frac{1}{\gamma}\frac{\partial\eta}{\partial\varphi}\right)\right]\right\}_{r=r_0+\eta(\theta,\varphi;t)} . \tag{A12}$$

B. Pressure term H_c in the potential energy

Turn now to the pressure term H_c in the potential energy given by

$$H_c = p_0 \int_0^{2\pi} \int_0^{\pi} \int_0^{r_0+\eta(\theta,\varphi;t)} dr \; r^2 \sin\theta \, d\theta \, d\varphi . \tag{A13}$$

The first order variation in H_c due to a small variation in η is

$$\delta H_c^{\eta} = p_0 \int_0^{2\pi} \int_0^{\pi} \left(r^2 \delta\eta\right)_{r=r_0+\eta(\theta',\varphi';t)} \sin\theta' \, d\theta' \, d\varphi' . \tag{A14}$$

The variational derivative $\delta H_c / \delta\eta$ can be found using Eqs. (A11) and (A14). The result is

$$\frac{\delta H_c}{\delta\eta(\theta,\varphi;t)} = p_0 . \tag{A15}$$

The formulas for H_b, H_c, $\delta H_b / \delta\eta$ and $\delta H_c / \delta\eta$ found so far in this appendix are useful in studying the energy of the system and in explicitly representing Hamilton's canonical equations starting from the postulated Hamiltonian.

Turn next to the problem of deriving an explicit representation for the pressure term in Cauchy's integral of Euler's equation of motion. Let

p=pressure just inside the free surface of the liquid;

p_0=pressure just outside the free surface of the liquid.

The differential change in volume of the liquid that occurs when a surface element $d\vec{f}$ is displaced radially outward by an amount $\delta\eta(\theta,\varphi:t)$ is given by

$$d^3r = \left(d\vec{f}\right)\cdot\hat{r}\delta\eta(\theta,\varphi;t)\,. \tag{A16}$$

The work $\delta H_c'$ done by a non-system worker in producing this displacement is

$$\delta H_c' = -\int_0^{2\pi}\int_0^{\pi}\left[(p-p_0)(\delta\eta)r^2\right]_{r=r_0+\eta(\theta,\varphi;t)}\sin\theta\, d\theta\, d\varphi\ , \tag{A17}$$

where Eqs. (A5b) and (A6a) were used in evaluating $d\vec{f}\cdot\hat{r}$. With the aid of Eqs. (A11) and (A19), one finds

$$\frac{\delta H_c'}{\delta\eta(\theta,\varphi;t)} = -\left(p-p_0\right). \tag{A18}$$

If the surface displacement occurs under conditions of local mechanical equilibrium, the sum of the pressure term and the surface term must vanish, so that

$$\frac{\delta H_b}{\delta\eta(\theta,\varphi;t)} + \frac{\delta H_c'}{\delta\eta(\theta,\varphi;t)} = 0\,. \tag{A19}$$

After rearrangement of terms, one finds

$$p = p_0 + \alpha\left\{\frac{1}{r}\left(\frac{1}{\gamma}+\gamma\right) - \frac{1}{r^2\sin\theta}\left[\frac{\partial}{\partial\theta}\left(\frac{\sin\theta}{\gamma}\frac{\partial\eta}{\partial\theta}\right)+\frac{\partial}{\partial\varphi}\left(\frac{1}{\gamma}\frac{\partial\eta}{\partial\varphi}\right)\right]\right\}_{r=r_0+\eta(\theta,\varphi;t)}\,. \tag{A20}$$

The expression for p in Eq. (A20) can be substituted into Eq. (9a) to arrive at the dynamical boundary condition at the free surface in Eq. (9b).

APPENDIX B

Demonstration that L=1 terms in surface displacement imply displacement of the center of mass of an almost spherical drop

In this appendix it is demonstrated that if at least one $\ell = 1$ term appears in the surface displacement given by Eq. (55a), i.e. by

$$\eta(\theta,\varphi;t) = \sum_{\ell,m,s} z^s_{\ell,m}(t)\; C^s_{\ell,m}(\theta,\varphi)\,, \tag{B1}$$

then the center of mass of a drop that is spherical or nearly spherical is displaced from the origin of coordinates for the angles θ and φ.

The surface of the drop is at

$$\vec{r}(\theta,\varphi;t) = \hat{r}\, r(\theta,\varphi;t) \tag{B2}$$

where

$$r(\theta,\varphi;t) = r_0 + \eta(\theta,\varphi;t)\,. \tag{B3}$$

The center of mass of the drop is located at position $\vec{R}$ given by

$$\vec{R} = \frac{1}{M}\vec{A}\,, \tag{B4}$$

where the mass M of the drop is given by

$$M = \rho \int_0^{2\pi} \int_0^{\pi} \int_0^{r_0+\eta(\theta,\varphi;t)} r^2 \sin\theta\, dr d\theta\, d\phi \tag{B5a}$$

$$= \frac{\rho}{3} \int_0^{2\pi} \int_0^{\pi} \left[r_0^3 + 3r_0^2\eta + 3r_0\eta^2 + \eta^3 \right] \sin\theta\, d\theta\, d\varphi\,. \tag{B5b}$$

The term linear in η integrates to zero. Thru first order terms in η we have

$$M = \frac{4\pi}{3} r_0^3 \rho\,. \tag{B5c}$$

The vector $\vec{A}$ is given by

$$\vec{A} = \rho \int_0^{2\pi} \int_0^{\pi} \int_0^{r_0+\eta(\theta,\varphi;t)} \vec{r}\, r^2 \sin\theta\, dr\, d\theta\, d\varphi . \tag{B6}$$

To evaluate $\vec{A}$ first write

$$\vec{r} = r\hat{r}, \tag{B7a}$$

$$\hat{r} = \hat{x}\sin\theta\cos\varphi + \hat{y}\sin\theta\sin\varphi + \hat{z}\cos\theta \tag{B7b}$$

$$= -\sqrt{\frac{4\pi}{3}}\left[\hat{x}\, C_{1,1}^{1}(\theta,\varphi) + \hat{y} C_{\overline{1},1}^{-1}(\theta,\varphi) + \hat{z}\left(-\frac{1}{\sqrt{2}}\right) C_{1,0}^{1}(\theta,\varphi)\right]. \tag{B7c}$$

In the last step we have used Eqs. (57a) and (57b).

Now rewrite Eq. (B6) with the aid of (B7a)-(B7c). Then integrate over r and retain terms only thru first order in η. The result is

$$\vec{A} = -\rho\sqrt{\frac{4\pi}{3}} \int_0^{2\pi} \int_0^{\pi} \left[\hat{x}\, C_{1,1}^{1}(\theta,\varphi) + \hat{y} C_{\overline{1},1}^{-1}(\theta,\varphi) + \hat{z}\left(-\frac{1}{\sqrt{2}}\right) C_{1,0}^{1}\right] \\ \times \frac{1}{4}\left[r_0^4 + r_0^3 \eta(\theta,\varphi;t)\right] \sin\theta\, d\theta\, d\varphi . \tag{B8}$$

The term involving r_0^4 vanishes upon integration. The term involving η can be integrated with the aid of Eqs. (B1), (61), and (62a)-(62c). After integrating and summing over Kronecker deltas, one finds

$$\vec{A} = -\rho r_0^3 \sqrt{\frac{4\pi}{3}} \left[\hat{x}\, z_{1,1}^{1}(t) + \hat{y} z_{\overline{1},1}^{-1}(t) + \hat{z}\left(-\sqrt{2}\right) z_{1,0}^{1}(t)\right] . \tag{B9}$$

Next evaluate $\vec{R}$ using Eqs. (B4), (B6), and (B9). One finds that the center of mass vector of an almost spherical drop is given by

$$\vec{R} = -\sqrt{\frac{3}{4\pi}}\left[\hat{x}\, z_{1,1}^{1}(t) + \hat{y} z_{1,1}^{-1}(t) + \hat{z}\left(-\sqrt{2}\right) z_{1,0}^{1}\right]. \tag{B10}$$

This demonstrates that if at least one of the $\ell = 1$ coefficients in Eq. (B1) is different from zero, then the center of mass is displaced from the original origin of coordinates.

APPENDIX C

Scalar product of two vector spherical harmonics

Vector spherical harmonics can be constructed[19] by coupling scalar spherical harmonics to unit vectors in 3-dimensional space using quantum mechanical rules for combining direct products of angular momentum eigenfunctions, as follows:

$$\vec{Y}_{J,\ell}^{M}(\theta,\varphi) = \sum_{-1\le m\le 1} C(\ell,1,J;M-m,m)\ Y_{\ell,M-m}(\theta,\varphi)\hat{e}_m\,, \tag{C1}$$

where

$$\hat{e}_m^* = (-1)^m \hat{e}_{-m} \tag{C2a}$$

$$\hat{e}_m^* \cdot \hat{e}_{m'} = \delta_{m,m'}\,. \tag{C2b}$$

With the aid of these equations one can evaluate the scalar product of two vector spherical harmonics as indicated next:

$$\left(\vec{Y}_{J,\ell}^{M}(\theta,\varphi)\right)^{*} \cdot \vec{Y}_{J',\ell'}^{M'}(\theta,\varphi) = \sum_{-1\le m\le 1} C(\ell,1,J;M-m,m)\ C(\ell',1,J';M'-m,m) \tag{C3a}$$
$$\times\ Y_{\ell,(M-m)}^{*}(\theta,\varphi)\ Y_{\ell',(M'-m)}(\theta,\varphi).$$
$$= \sum_{-1\le m\le 1} (-1)^{M-m} C(\ell,1,J;M-m)\ C(\ell',1,J';M'-m,m) \tag{C3b}$$
$$\times\ Y_{\ell,(M-m)}(\theta,\varphi)\ Y_{\ell',(M'-m)}(\theta,\varphi).$$

Using Eqs. (82) and (C3b), one can obtain the results that we seek, which are Eq. (103) and a formula for the coefficients a_{L_1,M_1} that occur there. One finds

$$\left(\vec{Y}_{J,\ell}^{M}(\theta,\varphi)\right)^{*} \cdot \vec{Y}_{J',\ell'}^{M'}(\theta,\varphi) = \sum_{L_1,M_1} a_{L_1,M_1}(J,\ell,M;J',\ell',M')\ Y_{L_1,M_1}(\theta,\varphi), \tag{C4}$$

where

$$a_{L_1,M_1}(J,\ell,M;J',\ell',M') = \sum_{-1\le m\le 1}(-1)^{M-m} C(\ell,1,J;M-m,m) C(\ell',1,J';M'-m,m) \tag{C5}$$
$$\times\ b_{L_1,M_1}(\ell',M'-m;\ell,-(M-m))$$

and b_{L_1,M_1} can be evaluated using Eq. (84).

APPENDIX D

Solution of harmonic oscillator equation with harmonic driving term

This appendix is concerned with solving the differential equation in Eq. (116), viz.,

$$\ddot{\psi} + \omega^2 \psi = e^{i\omega' t} \tag{D1}$$

subject to the conditions

$$\psi(t) = 0 \quad for \quad t \le 0 \tag{D2}$$

$$\dot{\psi}(t) = 0 \quad for \quad t \le 0 . \tag{D3}$$

The solution can be found with the aid of Laplace transforms, as described next. Let $\tilde{\psi}(s)$ be the Laplace transform of $\psi(t)$, where

$$\tilde{\psi}(s) = \mathrm{L}\ \{\psi(t)\} = \int_0^\infty e^{-st} \psi(t) \ dt . \tag{D4}$$

The Laplace transform of Eq. (D1) is

$$\int_0^\infty e^{-st} \left[\ddot{\psi} + \omega^2 \psi \right] dt = \int_0^\infty e^{-st} e^{i\omega' t} dt . \tag{D5}$$

From a table of transforms, one obtains

$$\int_0^\infty e^{-st}\ddot{\psi}(t)\,dt = s^2\tilde{\psi}(s) - s\psi(0) - \dot{\psi}(0) \tag{D6}$$

$$\int_0^\infty e^{-st}e^{i\omega' t}\,dt = \frac{1}{s - i\omega'} \; . \tag{D7}$$

Using Eqs. (D1)-(D7), one finds

$$\tilde{\psi}(s) = \frac{1}{(s - i\omega')(s^2 + \omega^2)} \; . \tag{D8a}$$

Now consider the case where $\omega^2 \neq (\omega')^2$. Partial fractions can be used to convert Eq. (8a) to the form

$$\tilde{\psi}(s) = \frac{1}{\omega^2 - (\omega')^2}\left\{\frac{1}{s - i\omega'} - \frac{s}{s^2 + \omega^2} - \frac{i\omega'}{s^2 + \omega^2}\right\}. \tag{D8b}$$

The inverse transform of Eq. (D8b) can be readily constructed with the aid of a table. One finds

$$\psi(t) = \frac{1}{\omega^2 - (\omega')^2}\left\{e^{i\omega' t} - \cos\,\omega t - \frac{i\omega'}{\omega}\sin\,\omega t\right\} \tag{D9}$$

which can be expressed as

$$\psi(t)=\frac{1}{\omega^2-(\omega')^2}e^{i\omega' t}-\frac{1}{2\omega}\left(\frac{e^{i\omega t}}{\omega-\omega'}+\frac{e^{-i\omega t}}{\omega+\omega'}\right). \tag{D10}$$

Equation (D10) agrees with Eq. (117a), and this accomplishes one of our goals.

Turn next to the case where $\omega^2=(\omega')^2\neq 0$. In this case Eq. (D8a) can be written as

$$\tilde{\psi}(s)=\frac{1}{(s-i\omega')^2(s+i\omega')}\ . \tag{D11}$$

Using partial fractions, one can write Eq. (D11) as

$$\tilde{\psi}(s)=\frac{1}{4(\omega')^2}\left(-\frac{1}{s+i\omega'}+\frac{1}{s-i\omega'}\right)+\frac{1}{2i\omega'}\frac{1}{(s-i\omega')^2}\ . \tag{D12}$$

Using a table of inverse transforms, one obtains

$$\psi(t)=\frac{1}{4(\omega')^2}\left(-e^{-i\omega' t}+e^{i\omega' t}\right)+\frac{t}{2i\omega'}e^{i\omega' t}\ . \tag{D13}$$

For each of the cases $\omega=\omega'$ and and $\omega=-\omega'$, Eq. (D13) can be expressed as

$$\psi(t)=\frac{t}{2i\omega'}e^{i\omega' t}+\frac{1}{4\omega\omega'}\left(e^{i\omega t}-e^{-i\omega t}\right). \tag{D14}$$

Equation (D14) agrees with Eq. (117b), and so the second goal of this appendix has been reached.

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